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JOURNEYS in SCIENCE
SMALL STEPS--GREAT STRIDES

Edited by
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Preface

THIS VOLUME is a collection of articles which are based upon lectures presented at the Twelfth Science Seminar of the Air Force Office of Scientific Research, held at Albuquerque, New Mexico, in June 1967. The title of this book, JOURNEYS IN SCIENCE: SMALL STEPS—GREAT STRIDES, which was also the theme of the seminar, evolved from an idea expressed by Dr. William J. Price, Executive Director of AFOSR, in his introduction to the book, SCIENCE IN THE SIXTIES, a publication resulting from the Tenth Anniversary AFOSR Science Seminar in 1965. Dr. Price said, "What has emerged vividly is the interlocking strength of seemingly disparate avenues in the sciences today, and the clear indication that great progress is often made in small steps occurring in a variety of ways."

This book, strictly speaking, is not a publication of the proceedings of the seminar. Instead of consisting of verbatim transcripts of the lectures, it is a collection of articles which, in almost every instance, were written especially for the book. Most of the chapters represent condensed versions of the two lectures presented by each of the distinguished researchers appearing on the program. The state of the art in many scientific areas embraced by the interests of the Air Force Office of Scientific Research is clearly delineated in this collection.

The seminar, sponsored by AFOSR, was held in cooperation with the University of New Mexico and the Air Force Special Weapons Center.

I am greatly indebted to my wife, Rena, who assisted in reading the manuscripts; to Mrs. Opal Broome, Miss Barbara Malecki and Mrs. Wanda Climenhaga for assistance in reading proof and to Cadet John C. Frost, of the United States Air Force Academy, who made the drawings which illustrate Chapter IX.

D. L. A.

Arlington, Virginia
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Contents

I. Research on Research

DEREK J. DE SOLLA PRICE

DEDICATION

I should like to dedicate this lecture to the memory of that distinguished friend and Yale colleague, our "flying philosopher," Norwood Russell Hanson, who was to have given this keynote address. Russ and I were first together in Cambridge, England from 1952 through 1956 where we enjoyed to the hilt all the fun of pioneering in our respective lobes of the dual disciplines of History and Philosophy of Science. His death on 18 April 1967 in the crash of his Grumman F8F Bearcat plane was an event that all his friends had anticipated daily, much to his delight, for the past several years. The rehearsal does not, however, take the sting from the way in which we now miss this strangely versatile and creative man.

The subject for these small but cumulatively significant steps of scientific inquiry which we shall report is somewhat unusual. Most scientists study the physical world around them and its animate and inanimate behaviors—they are biologists and physicists, chemists and astronomers. The target of our curiosity is the nature of science itself and the behavior of the scientist in seeking it out. Why and how does the scientist do what he does? Why does science "come out" in just the way it does and not in some other way? What, in short,

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to use the scientists' own metaphor, makes science tick? How is it, indeed, that a succession of single steps can cumulate in scientific knowledge in a way that seems to reach farther and faster than non-scientific knowledge?

As a preamble, because of the special character of this research, it is necessary to cast a glance at what George Sarton once called the Parascientific Professions, the academic specialties of the history and the philosophy of science, together with the more recent accretions of sociology and economics of science, the psychology of scientists (and their psychiatry, insofar as that is special). Perhaps one ought also to include the fields of research management studies and science policy studies, both waxing more and more diverse and ever stronger with each growth of science in big business and bigger government.

The whole complex, together with the sort of material I shall display today, is sometimes called "science of science"; I dislike the name, hence the experiment with the title of this chapter. Nevertheless, reduplication of names seems to have gone out of fashion in science since the day when Galileo Galilei was christened. My own predilection for this type of intellectual labor, rather than any other, stems from a desire to see all parts of the research fronts of all of the sciences; while wooing the muse of history, I never quite managed however to forget that I had been a physicist. I, therefore, have always had a strong temptation to try to understand the workings of science, not in the usual humanistic way, but with the hard and precise quantitative and mathematical tools of the physicists' modes of thought and comprehension.

The urge is one thing, but the means of fulfilling it is another. Quantitative thought is quite impossible without a goodly stock of data. When Bernal wrote on the social organization of science in the late 1930's, there were hardly any accurate data at his command to tell how many scientists there were and how much of what science was being done in what country. Now, well into the age of atoms and space, and thanks to the governmental pre-occupation for accounting for its funding and manpower, we have compendious statistics from several countries including the prodigious blocks of data, machine handled and issued in bulk by all the Federal agencies of the United States Government. Then again, thanks to the need to look after the giant rise of scientific literature and the swol-

len scientific societies and their journals we have, often quite incidentally to the main function of management, a storehouse of data that never was available anywhere until about a decade ago.

In some respects the supply of data is an embarrassment here as it has been in meteorology. It takes a great deal of data to make even a weak theory, and it takes a great deal of theory before you can explain any small quantity of the data with a sufficient understanding to make little predictions. The situation also resembles rather strongly the stage of astronomy in the period of Tycho Brahe, which is very familiar to the historian of science as a key case study. A mass of data has been accumulated; we have begun to make the empirical generalizations and projections comparable to the Kepler Laws of planetary motion. The present stage must be a continuation of both efforts together with the first faltering steps towards those Newtonian Laws that will explain exactly why the Kepler motions behave as they do.

To develop an understanding of this sort is difficult. It involves the necessity of being not only literate but also numerate. We shall have to think in numbers. It is made all the more difficult by the fact that the scientists here become their own experimental animals in a Haldane-like fashion, and they suspect violently, sometimes almost psychotically, all attempts to evaluate them and their work in the way that they evaluate the universe. In this case the suspicions are at least partially reasonable, for the basic methodology bears a nasty resemblance to what I am told is the Texas method of hog-weighing. A hog is put at one end of a seesaw board; stones are piled on the other end till the board balances, then you guess the weight of the stones.

The procedure in this work has been to use headcounts of scientific manpower, published papers and journals, patents and money, and then to suppose that these quantities measure in some mysterious way the "size" of science. It is perhaps not so bad as it sounds. Most of the measurements show a very strong statistical regularity and they obey with remarkable precision their equivalents of the Kepler Laws. Science, measured in this sort of way, loose though it seems, is much more regular in its behavior than most other measurable characteristics of our civilization. Science is, so to speak, a much more regular thing in its behavior than are people.

Perhaps the best example of this regularity is to be seen in the

First Law of Research on Research (R on R). This law states that if you measure the size of science as a function of time, the progress is seen to be a regular exponential increase, holding for periods as long as 200 years, and maintaining a rate of compound interest growth of the order of a doubling every 10 to 15 years, a multiplication by a factor of 10 two or three times during each century. One gets about the same rate whether you count men, or scientific journals, or the papers published in them. Rates vary only a little from field to field of science, from country to country.

To push this example to the next stage, beyond being merely empirical, we must ask two sorts of questions. Firstly, why i· the regular growth exponential and how can one compute its rate of growth from other parameters? Secondly, given this law of growth, what other consequences flow from it, and how will it relate to other aspects of science which may be measurable? The first question, demanding a knowledge of any underlying mechanism, has an obvious lead: to a mathematician it is obvious that exponential growth follows as a solution of the differential equation, $dx/dt = kt$; that is, the situation holds where the rate of growth is somehow held in constant proportion to the size of the population attained. Scientists beget more scientists and old knowledge brings forth new knowledge at a constant rate. This is true, it seems, for all knowledge and all types of people, but for science and scientists there appears, as we shall show later, to be some special research front structure that lends to this activity a growth rate far in excess of all the other activities of mankind.

The second question, asking the consequences of this mode of growth, has some rather famous answers. Among these consequences are the facts that most of the scientists that have ever lived are alive now, and that most of these scientists alive happen to be rather young persons. It is quite easy to prove these alarming facts. Alive now there exist, roughly speaking, all scientists who were born between 20 and 65 years ago. At a minimal estimate this is about three of our periods during which science doubles in size. Therefore, for every man born before the first of these periods there will be one more from the first doubling, two from the second and four from the last. Thus, out of every eight scientists that have ever lived seven are alive right now, four of them being fellows who have come into the business during the last fifteen years. It

follows then that $87\frac{1}{2}\%$ of all the scientists who have ever been (say, 90%) are alive now, and of these more than half have been in the occupation for less than 12 years.

Although the argument is so simple, the facts give a rather deep explanation of some of the more pervasive characteristics of science as noted by historians and sociologists. Science has a persistent quality of immediacy and juvenility. Not only is it now true that most of the scientists are young, and half of all we know has been found out in the last decade or so, but this has always been true. Since it is a consequence of the exponential growth that has held sway for the last two or even three centuries, Ben Franklin and perhaps Isaac Newton could have said just the same things about how much is new and how much science was a product of their day and age as it never had been before. Science runs so much faster than people, so much more rapidly than civilization.

This is by no means as acute as the situation is, in fact. Men come into the field and new ideas come on the scene; some men leave the field and some ideas disappear to the limbos of rejection and obsolescence. By any reasonable measure it can be shown that the death rate is of the same order of magnitude as that of overall increase. Since the general growth rate is of the order of 7% per annum, it follows that this comes about through a birth rate of something like 15% per annum combined with a death rate that is also 7% or 8% per annum. The situation is rather similar to that of a very primitive village where all the women are giving birth almost every year, but the state of medicine is such that an appreciable segment of the population, adults as well as children, dies every year. The result is that the village contains a quite small core of adults who provide the labor force and who have happened to have survived so long, together with a large bulk of young children who are, so to speak, just passing through this existence.

It turns out that in such a village, or to come back to our scientists, in the population of science, most of the practitioners are newly at it, but about half the man-years of work are due to that very small stable bunch of individuals who happened to have survived the quite enormous mortality. In hard numbers, whichever way measurements are taken, the size of the hard core, responsible for about half the man-years of work, half the papers, turns out to be about the square root of the size of the total population. The large number of

children "just passing through" have their analogues in science too. In this age of heavy financial dependence of young scientists on old, the young become the assistants and collaborators of the senior men, and it is this process that accounts for the rapidly-increasing amount of collaborative authorship in scientific papers. In 1870 it was very rare for a scientific paper to have a pair of authors instead of just one claimant of intellectual property. By 1970 we shall be well on the way to complete extinction of the single-author paper in the most heavily pressed and financed scientific fields and we shall even be moving towards the phenomenon of an almost infinite number of authors on each of the almost infinite number of papers. All the subsidiary authors, just passing through, are part of the rat-race of modern science; their names swell the list of *American Men of Science* and in the next edition, so many of the names will be gone, others will have taken their place, and only the small stable square-root population will remain as a core of "known" people.

Again in the spirit of our inquiry there are two ways of proceeding in this case; firstly we can ask for a "Newton's Law" governing the rat-race, secondly we can ask for the practical consequences over and above those that have just been mentioned. Perhaps the chief of these consequences is that one can quite easily show by investigating the minor characters, that collaboration is not a pooling of talents to produce better work or different work; in most cases it must be a device for getting papers out of people who essentially seem to have less than a half paper in them. The number of such people is far in excess of twice those with a whole paper in them; so by using this lesser quality manpower, one can more than double the output of scientific work. All that is needed is a sufficiency of able and senior people to lead the teams and a sufficiency of incentive to make the large numbers of lesser men aspire readily for the chance of surviving to become one of the great and old.

To cool the situation a little I strongly suggest we determine to award credit for authorship at the rate of $1/n$ of a point for each of n authors of such a collaborative paper. It follows quite well from our laws that they do not, on an average, deserve more than this. Certainly they do not deserve a whole point each.

The exact law from which this all comes, the law of the distribution of quality among men, papers, journals, institutions, etc., might

be called the Second Law of Research on Research. It turns out that the distribution in all cases is rather similar to the Pareto Law of the distribution of economic incomes within a country. It is also similar to the Zipf law which governs the sizes of cities within a territory, or the length of words or sentences in a sample of language. All of these laws are mathematical approximations that are roughly equivalent one to the other, being lognormal or almost inverse square law distributions. They are very assymetrical and quite unlike the usual Gaussian type distributions that are of universal applicability to such entities as the height and weight and intelligence of large populations.

One way of looking at such distributions is to compare them with the case of populations of soap bubbles. Such bubbles have the curious property that when two meet and come into interaction, the little soap bubble blows up the big one. In most other cases of physical interaction it is the big that gives to the small so that the universe slowly evens out on its way to that great heat death of the entropy law. With soap bubbles and with scientists the sign of the interaction is the other way round, and the statistics of the population tend to a condition in which most of the space is occupied by a very few very large bubbles. Correspondingly, one gets most of the scientific work done by a very few very clever and productive scientists, though a very large number of lesser characters are striving for this eminence and dying like flies on the way.

To be a little more exact about this distribution, it happens numerically for men as for institutions that the chance of doubling the size of the achievement is uniformly about one in four, no matter what the size already attained. The chance that you will publish ten more papers if you already have published ten is about 1 in 4: the chance of doing a second paper if you have only so far done your first is 1 in 4. The mortality is always the remaining three chances out of four: always the mortality is due to this soap-bubble principle: the more eminent and the more experienced tend to get results before those of less degree. Science happens to be such that there seems, after all, to be only the one world to discover. If the constant is discovered by Planck there cannot be this discovery again: it has been made. If Beethoven had not existed, other men would have written quite different symphonies; Beethoven's private property is unmistakable. If Planck, however, had not made this

particular discovery, somebody else would have had to have made it, and the indications are to the historian of modern science that the somebody else would have made the discovery rather quickly. Most discoveries worth making are indeed made by many more than a single person so that the syndrome of disputed priority and subsequent contest for recognition is one of the most common within history. Merton has considered this soap-bubble type of action from the standpoint of the sociologist and for him it has become the Biblical Matthew Principle, "unto him that hath shall be given, and unto him that hath not shall be taken away, even that which he hath." Science is a quite grim battleground.

At this point we must backtrack a little to consider the basic mechanism by which science cumulates and is enabled to grow exponentially at a rate so much more surefooted and rapid than mere non-science. Our knowledge about this comes as an incidental benefit from the production of the *Science Citation Index,* a quarterly and annual ongoing publication in which you can look up any paper that has ever been published in the past and find out who, if anybody, has been citing it as a reference in their more recently published work in more than 1000 of the world's leading scientific journals in all fields. The great advantage, of course, is that this is the only sort of index that runs forward in time instead of backward to older and older material; it is also the only sort of subject matter categorization that does not let a cataloguer, librarian, or other "expert" intervene between the generator and the seeker of information.

Each of these annual citation indexes carries data on millions of references and we have been most fortunate in being able to use the results of various machine sortings and countings to determine in just what sort of way papers are tied together by their practice of citing each other. It turns out that there are two main variables involved The first of these is the amount of citation, the number of references cited by each paper on the average. Throughout the whole of science the general average is of the order of about ten references per paper. If any field carries far less than this, say one or two or none at all, the presumption is that this is not an area where papers are built on the foundation of previous papers. Quite often in technological or professional magazines one finds this sort of article, serving for news value or some other purpose, rather than

that of making a contribution to scholarship. Then again if there are many more than ten references, say thirty or forty, the presumption must be that this again is a beast of a different kind, probably a review type of paper summarizing all recent previous work in a given field.

A second variable concerns the way in which those references that exist actually tie the new papers to the old. For highly non-scientific fields it happens that any piece of material that has ever been published has about an equal chance of being cited, no matter what its date. Papers of all dates may be very good or very bad, but the mix seems about the same for all dates, ancient and modern. With science it is quite different: as Heisenberg's father once said, each paper comes with only three months' guarantee. Very recent papers or even those so new that they have hardly been circulated in preprint form, let alone published, are often much more valuable, more frequently cited than papers which have been around for enough time so that the good juice has already been squeezed from them and everybody knows the material anyway. For normal scientific fields the citations to papers published during the last five years is about 30%. For the hardest scientific disciplines such as molecular biology and theoretical high energy physics, where scientists are treading hard on each others heels (and heads), the proportion may rise to as high as 70%. For a few peculiarly archival fields such as zoological and botanical taxonomy, where older archetypes are preferred, the proportion of recent work may be as low as 15%.

The hardest science grows from a very thin skin, whereas ordinary scholarship grows from the body of its knowledge. By growing from the skin alone, the proliferation is concentratedly attached to a few papers instead of being diffusely related to so many. Unfortunately, we do not have any decent mathematical formulation of the statistical properties of networks of this sort, and only in a bumbling way have we been able to divine and guess the various interrelations. In a few cases, where a sample subject can be isolated and is small enough to consider in detail, one can draw a complete diagram of the structure. It turns out from this that science has a research front something like forty papers deep; anything older than this tends to be packed down into reviews and then into textbooks—if it lasts at all. Most contributions exist only for the purpose of en-

abling some other small and current advance to be made—once this has been made the paper that spurred it becomes useless. It is not intended to be packed down and become archival, even though the strong superstition of scientists is that publication is somehow a sacred duty in order to read the contribution into this perpetual and immortal archive.

The mythology of an archive is something that runs deep in the life of science and is worthy of the hardest analysis. It is a most intriguing paradox that the scientist secures the maximum in private intellectual property by the device of the most open publication. Publication is the key to it. Papers and journals exist as a medium for rapid publication, preferably together with the conferring of an aura of status and approval through stature of the journal and discretion of the esteemed editors. There is little evidence that such journals and papers are actually read as a means of transmitting the scientific information so printed. That knowledge has already been sent around the research front by private circulation and informal means. The open publication is, of course, scanned to see who got what in this week, and it is naturally the means whereby graduate students can, by hard reading, reach the research front. It would be no good at all if science was able to run so fast that embryonic new scientists could never catch up with the advancing front.

Scientists, it seems, are those who are highly motivated to publish but not to read. Interestingly enough it is a totally different situation in the technologies. One might almost define a technology as a field where the chief intended product is an object, a manufacture, a process, a chemical, rather than a paper. Publication there might be (probably in simulation of the approved sciences) but it is readily seen to be epiphenomenal. Technologists do not want to publish usefully—there is no tradition of giving one's competition a useful lead—but they want very much to read in case somebody else has let slip a lead out of which they think they should be able to get some useful advance of practical significance. For the most part, such literature crisis as is often discussed is an artificial construct on the part of technologists who believe there is some sort of useful scientific information archive to which they have only a most difficult access. In fact the sort of material they want is not published

at all. The scientist is concerned with the publication of quite different material.

The most surprising revelation from such an analysis is that if one defines science and technology in these sorts of terms instead of the rather weak and naïve definition by intent that we usually use, it becomes most perplexing to analyze any relation that may exist between various parts of science and various parts of technology. So far as I have been able to take this analysis, science and technology are separate and almost independent cumulations. Old science breeds new science at constant rate through the medium of a knitted network of literature, and analogously old technology breeds through an interwoven net which consists not of an embodied literature, but of a knowhow that is only partially recorded in such things as patents and advertisements and the trade catalogues. Between the two networks there is, I think, what the physicist would call a "weak interaction", just sufficient to keep the two cumulations in step. Only atypically, in those rare traumatic incidents that become legend, like transistors and penicillin, does a piece of new science immediately or quickly give rise to a new advance in technology. For the most part, transfer from one to the other is carried in people who migrate from one side to the other in course of training or shift of jobs. At all events, it is utterly wrong to conceive of technology as being equivalent to applied science.

Indeed, in the matter of interaction between science and technology there is even greater complication. Some technology may not be related to science at all. Quite a lot of steam engine technology had nothing whatsoever to do with thermodynamics, and quite a lot of engineering goes like the invention of zip-fasteners and safety pins, through a home inventor or bicycle shop mechanic mechanism rather than through anything that could be recognized as "scientific" training. A very large part of the activity which is now traditionally called "development" and even "applied research" consists of what a manufacturer must do in order to try out and start making a new product. If such trying and starting uses any considerable quantity of scientific training, I suppose it should be called research, but if it runs quite close to manufacture, say as a pilot operation, it would seem better to include it as part of the process and necessary expense of production.

Some light on this central problem of the science of science may be had from some new research into the economics of science, here presented for the first time. I might add that this seems to be a good case of simultaneous discovery; several of us in several countries have hit upon much the same idea at the same time and, naturally, each of us feels that he is the true begetter and originator of the notion. We have all found a rather interesting regularity in a sort of data where regularities worth talking about have never been noted.

To put it in a nutshell, the issue is why the United States, for example, seems to produce about one third of all the world's physics and chemistry and most other sciences. Why is the share one third and not, say, about 6% which is its share of the world population, or 90%, or anything else? About a year ago one of the most famous grand old men of science in the USSR asked why the USSR was only about half the size of USA in scientific output of papers in all fields even though the statistics showed that the number of scientists in the two countries were roughly comparable and certainly not different by a factor of two. Why do Canada and India each publish about 2% of the world's science though India has a population 25 times greater than that of Canada? Not only do they publish about equally, but also their governmental and professional societies and organizations are of about the same size, structure and complexity, though of course different as are the countries in their politics and philosophies. Science has to be universal. There is only one game of physics to play and it makes little difference whether one approaches it as one religion or another, one politics or another.

Our discovery is simply that one gets a feeling that the size of each country's scientific effort is proportional, not merely to the size of its population, but also to something like the per capita wealth of that nation. Now, if one multiplies population by wealth per unit population one gets total wealth. We come, therefore, to the conclusion that at any instant of time the several scientific outputs of the nations of the world should be proportional to the Gross National Product (or something like it) of each nation. It is not worth refining the theory to see which of the several definitions of GNP, personal income etc., should be used—they are all equivalent to the extent of this rough computation. To put the result in the terms in which we posed the question, the USA is about one third

of the world's science because it is also about one third of the world's wealth.

In Table I we have set the best available figures for the numbers of papers in *Physics Abstracts* and *Chemical Abstracts* alongside those for the wealth and the populations of the chief countries of the world that play a significantly large role in publishing the science that is our common stock. It will readily be observed without

TABLE I
NATIONAL BROWNIE POINTS

Country	Percentages of World Total			
	Share of GNP 1964	Share of Phys. Abstr. 1961	Share of Chem. Abstr. 1965	Share of Population 1964
(i) Larger participants:—				
USA	32.8	31.6	28.5	5.9
USSR	15.6	15.6	20.7	7.0
W. Germany	5.2	} 6.2 {	6.3	1.8
E. Germany	0.8		2.2	0.5
UK	4.8	13.6*	6.7	1.6
France	4.5	6.3	4.5	1.4
Japan	3.6	7.8	7.3	2.9
Italy	2.6	3.4	2.7	1.5
Canada	2.2	1.1	2.0	0.6
India	2.2	1.8	2.2	14.4
Poland	1.6	1.5	2.9	0.9
Australia	1.1	0.5	1.2	0.3
Romania	1.0	0.6	0.9	0.5
Spain	0.9	0.2	0.4	1.0
Sweden	0.9	0.7	0.9	0.2
Netherlands	0.9	5.2*	0.8	0.4
Belgium	0.8	0.3	0.6	0.3
Czechoslovakia	0.7	0.9	1.6	0.4
Switzerland	0.7	1.0	1.0	0.2
Hungary	0.5	0.5	1.0	0.3
Austria	0.4	0.2	0.5	0.2
Bulgaria	0.4	0.2	0.5	0.2
(ii) Smaller participants and non-participants:—				
All other countries	15.8	0.8	4.6	57.5

* Note: Data known to be swollen because of one or more large international journals published from this nation.

any apparatus of statistical correlations that most countries show pretty good agreement between their shares of wealth and those of the pure sciences. The United Kingdom and the Netherlands have distorted figures for Physics because they do much more than their share of publishing great and important international journals in this field; though published from those countries, their contributors are from elsewhere.

Apart from this the contributions to science of Japan and the U.K. are both rather high, or to look at the reverse of that coin, their GNP's are uncommonly low for nations that scientific. Spain, on the other hand, does much less science than it should. Apart from these, the shares, to this order of accuracy, show excellent agreement with expectation. Outside the club of significant scientific nations there stands 57% of the world's population with nearly 16% of the wealth but only less than 5% of the science—and among these countries are such places as Denmark which is highly active but too small to show on our table. Missing from the list are great blocks such as Latin America that should have 3.7% of the science, China which should be 3.5% of the world, Africa at 1.7% and the Near East which should publish about 1%—these alone amounting to 10% of the residual 16% of wealth.

From more detailed accounts of the distribution of special fields among the countries of the world we come to the conclusion that, to a first approximation, all the pure science areas are distributed fairly normally as we have seen for physics and chemistry. There is a sort of equipartition; if a country has 1% of the world's physics it probably has 1% of the pure mathematics and 1% of the bio-chemistry. The balance between the sciences seems to change only slightly from country to country, and only very gradually with time within any one country. For the applied sciences it seems very different; highly agricultural nations have a lot of agricultural science and those without agriculture do not. It is similar for the other applied sciences such as mining, engineering construction, airplane manufacture, etc. The applied science or technology activity of each nation—whatever one calls it—depends on very much more than the wealth of the country. Each nation seems quite rightly to make its own decisions to deploy its useful labor into regions that are valuable to it but not necessarily for other lands. Science again

satisfies an international constraint; either you play the game and conform with the world deployment or you tend not to play at all.

The implications of this tentative equality between scientific output and economic wealth of nations is considerable; it may be of the greatest importance to all matters of general science policy. On the world scale there are published each year about 700,000 scientific papers, by about the same number of authors, and the GNP of the whole world is about $2 x 10^{12}. There must be therefore about three million dollars of GNP of each country for every scientific paper in that country, or about the same for each author. If we reckon an average salary at about $10,000 per annum and an equal amount for the overhead in plant and equipment, secretaries and paper clips, the cost of the scientists comes out to be just 0.7% of the GNP of every country that is participating in modern science. An alternative route to the same result is to take the agreed and standard figure for the cost of research index—at about $20,000 per published paper. It should be noted however that both computations and the final result of 0.7% of GNP refer only to the present situation. The change with time may be quite rapid. Both scientific salaries and the GNP (or personal income) of countries tend to rise together at a rate that is typically about 4% per annum. The amount of scientific work, in terms of numbers of papers and of scientists is rising much more rapidly, typically with a doubling every ten years. It follows then that simply to maintain the status quo, each country must double the percentage spent on pure science from its GNP in every decade. Though we have a figure of 0.7% now, it was only 0.3% in 1956 and it will be 1.5% in 1976 and 12% in 2006, assuming similar conditions to hold.

Such conditions cannot of course hold indefinitely, and indeed they begin to break down quite rapidly because in the most developed countries the 0.7% we have just computed is only a small fraction—about one fifth or one quarter of the total expenditure on what is traditionally known as R & D, research and development. Since we have already computed this 0.7% as the amount needed (give or take a factor of two perhaps) to support the pure sciences (and perhaps any "pure technologies"), it follows that the other two thirds or three quarters of the national expenditure must be going to buy technology, presumably to buy that sort of technology

that can be billed as "research" with all the aura of science rather than simple manufacture. If we were to spend 12% of the GNP on pure sciences, we should, to preserve the same balance be spending a total of half to two thirds of the GNP on R & D—an almost inconceivable amount.

At present the total USA expenditure on R and D is of the order of 3.5% of the GNP, and it was 1.4% a decade ago. The figures are thus running about five times what we have computed for the pure component of universal scientific knowledge, the paper-producing industry alone. There is indeed good reason why the maximum deployment of scientists outside the knowledge industry should be, at most, of order of magnitude three or four times those whom we have counted as pure scientists. Traditionally in the sciences only about 20% of the Ph.D. graduates are recycled so as to become college teachers. In the humanities the conventional figure is around 80 or 90% so that there is in those fields very little spare output. The chief service to the community being performed by humanists is the education of the young at the undergraduate level; at the graduate level the principal activity is reproducing their own kind, teaching students to become teachers to train more students in an endless cycle. In the sciences the surplus exceeds the needs of reproduction by a factor of four.

One rough way of accounting for this is to make use of the known rate of exponential growth to determine the needs in self-reproduction. For every active 100 scientists it is necessary to train about seven new ones each year; assuming that this is about a four year process there will therefore be about 30 students in residence at any time, and considering the national average it will take a supply of about 20 teachers at the most to care for them. This leaves some 80 active scientists who need not be teachers, and the ratio is about 4 to 1 as expected.

Of course, the ratio is only valid if the active scientists are in the same sort of field of science whether they teach or serve the community by delivering some other service or product. Thus it is possible to use in physics industries about four times the manpower of physics teachers, and in chemistry a similar proportion, etc. One cannot just decide to have a very large deployment of labor in, let us say, agricultural researches, for the manpower available is only four times that of the pure science in the related

areas of biology, and the amount of biological research, like those of the other sciences, is a fixed amount in equilibrium each with the other and all with the wealth of the country.

It would seem that in the smaller and less developed countries, the current total expenditure on science can be the calculated 1% as a minimum. Any expenditure beyond that, up to a possibility of about a factor of four beyond that, implies the support of the associated technologies. It is, of course, a vital question, but one on which there has not even been speculation to determine whether the wealth of countries is due to their attention to this deployment in technology, or whether only the amount supported is due to the wealth. At all events we know now that there tends to be a good correlation, so it is reasonable to suppose as a matter of policy that each country should spend about 0.7% of its GNP on the support of those whose business it is to engage in the knowledge industry and publish scientific papers. They may then spend up to four times as much in any sectors they choose where the trained manpower can be deployed in technologies useful to the particular needs and industries of the country concerned.

As a further trial of this important new principle, I present here, also for the first time, a set of calculations based on the wealth (measured in total personal income) and the scientific manpower in various fields (taken from the National Register for 1964 and 1966 of all of the separate states of the USA). In Table II we show a computer printout result which sets the states in order of the share of manpower per unit share of wealth. According to the theory a "normal" state should have this ratio about equal to unity, and indeed 15 of the 51 states come within 10% of unity, and a total of 34 are within 25% of unity—this is very good agreement, given the random noise expected in this. Only eight states have abnormally high ratios, and for each of them there seems a good explanation why there should be a much larger scientific population than one might expect. The District of Columbia is obviously artificial in its structure, Delaware has several well-known large chemical companies, New Mexico and Massachusetts are known to have abnormally large holdings in physics, Maryland has the National Institutes of Health, etc. At the other end of the scale are the educationally depressed states such as Arkansas, Mississippi, Kentucky and Georgia; this calculation shows that they have about

TABLE II

% Sc. manpower / % Personal inc.	State	Fields Overconcentrated	Fields Underconcentrated
5.61	D.C.	—economics ↑statistics	—chemistry
3.22	Delaware	—chemistry	
2.02	New Mexico	↑physics	
1.75	Wyoming	↑agriculture ↓earth sc.	
1.70	Colorado	↑agriculture ↓earth sc. ↑meteorology	—chemistry
1.60	Maryland	—biology	
1.50	Utah	—agriculture	
1.40	Massachusetts	—physics	—agriculture —earth sc.
1.25	Alaska	↑agriculture —earth sc. ↑meteorology	
1.25	Oklahoma	—earth sc.	
1.23	Idaho	↑agriculture	
1.22	Montana	↑agriculture ↓earth sc.	
1.21	New Jersey	—chemistry	—agriculture —earth sc. —biology
1.14	Arizona	↓agriculture	
1.09	Washington	↑agriculture	
1.07	Louisiana	↑earth sc.	
1.03	Oregon	↑agriculture	
1.01	California	↓physics ↓mathematics	—chemistry
1.00	Texas	—earth sc.	
1.00	Vermont		
0.99	Connecticut		
0.99	New York	—psychology ↓economics	↑agriculture —earth sc.
0.97	Tennessee		
0.97	Minnesota		
0.96	Pennsylvania	↓chemistry	—earth sc.
0.94	New Hampshire		
0.92	Hawaii	↑meteorology	
0.90	Virginia	↑mathematics	
0.90	West Virginia	↓chemistry	
0.85	Wisconsin		
0.83	Rhode Island		
0.82	Ohio	—chemistry	
0.80	Illinois	—chemistry	—earth sc.
0.00	Kansas		
0.79	North Dakota	↓agriculture	
0.79	Missouri		
0.79	Iowa		
0.79	Indiana		
0.78	Nevada	↑agriculture	
0.78	South Dakota	—agriculture	
0.76	Michigan		
0.76	North Carolina	↓biology	

(Left margin groupings: rows 5.61 through 1.40 labeled "more scientists than expected"; rows 1.25 through 0.76 labeled "Normal complement of scientific manpower in population")

TABLE II (continued)

$\frac{\% \text{ Sc. manpower}}{\% \text{ Personal inc.}}$	State	Fields Overconcentrated	Fields Underconcentrated
0.69	Nebraska		
0.69	Maine	↑agriculture	
0.67	Alabama	—agriculture	
0.64	Florida	—meteorology	
0.61	South Carolina	—agriculture	
0.61	Georgia	↑agriculture	
0.60	Kentucky		
0.57	Mississippi	↓earth sc. —agriculture	
0.53	Arkansas	—agriculture	

(left margin, bracketed:) fewer scientists than expected

↑maldistribution increases
—maldistribution constant } from 1964 to 1966.
↓maldistribution decreases

one half of the scientific activity they deserve and need for education and development. For more detail in this table we have taken each field in turn and measured the amount (a Chi value) by which it deviates from what one might expect if each science was distributed in the same proportions as the total scientific manpower. Only the largest and most significant deviations have been indicated, but as a matter of interest we have computed figures for both 1964 and 1966 and indicated on the table whenever there has been a considerable shift with time.

To set the same information in another way we have taken a field presentation in Table III; this shews immediately that it is

TABLE III

WHERE THE SCIENTIFIC ACTION IS
Large positive Chi values of (actual—expected) figures for scientific manpower.
Source: National Register 1966.

Agriculture		Earth Sc.		Meteorology		Chemistry	
Oregon	54.3	Texas	83.4	Foreign	43.1	New Jersey	44.0
Idaho	37.4	Louisiana	54.4	Guam	28.0	Delaware	35.8
Montana	36.2	Oklahoma	50.8	Alaska	19.8	Ohio	24.4
Washington	24.5	Colorado	40.7	Hawaii	15.4	Pennsylvania	19.9
Alaska	20.7	Foreign	35.2	Florida	12.2	W. Virginia	13.4
Arkansas	19.7	Wyoming	28.5	Colorado	11.4	Illinois	11.7
Georgia	19.4	Alaska	15.1				
Wyoming	19.1	Montana	11.6				
S. Dakota	19.1	Mississippi	11.3				
N. Dakota	13.7						

TABLE III (continued)

Agriculture		Earth Sc.	Meteorology	Chemistry
S. Carolina	13.3			
Utah	12.9			
Maine	12.2			
Nevada	10.9			
Alabama	10.5			
Arizona	10.1			
Colorado	10.1			

All Else		
District of Columbia	Economics	41.4
New Mexico	Physics	24.8
District of Columbia	Statistics	24.4
Maryland	Biology	23.5
Massachusetts	Physics	23.4
California	Mathematics	22.8
California	Physics	20.5
New York	Psychology	19.3
Foreign	Linguistics	18.8
Foreign	Anthropology	14.3
Virginia	Mathematics	13.7
New York	Economics	10.7
N. Carolina	Biology	10.7

the useful technologies that vary sharply from state to state according to their particular industries and needs. The exact sciences have very little maldistribution and those that exist all seem reasonable in view of the existence of special large laboratories and institutes.

The program is now being extended to take other computer methods for evaluating the statistical distributions of science, measured in many different ways, among the states and among the nations of the world. It might not tell one exactly what to do in science policy or how to decide whether a particular state should spend a certain amount of money on this or that activity, but it does represent a most useful monitoring system that can alert one to unintentional and accidental overconcentrations and underconcentrations. Our hopes are that as these and the other quantitative models develop, we shall more and more come to know just how science works in such a way that we can deploy our limited resources to the best advantage.

ACKNOWLEDGMENTS

II. Psycholinguistic Approaches to the Study of Communication

GEORGE A. MILLER

FOR MANY YEARS psychologists and communication engineers have collaborated to test and improve the quality of voice communication equipment. In this collaboration, the psychologist's major interest has frequently been in auditory perception, and his contribution to the team has generally been to determine which perceptual aspects of a voice signal must be faithfully transmitted if the message is to be intelligible to a receiver. Today this minor branch of applied science is reasonably well understood; if I wished to emphasize past accomplishments, I could review with some pride what we now know about the acoustic nature and psychological perception of speech.

It is more challenging, however, to tackle problems that are still unsolved. There, of course, is where our research attention must be focussed, and it is always more interesting to talk about what you are still puzzling over. So I intend here to discuss questions about some psychological matters that I believe are critically important for our understanding of the communication process, but whose answers are not yet completely clear.

Although we now understand a great deal about the communication of signals, the communication of meaning remains something of a mystery. When you begin to probe this mystery, you encounter immediately such enormously complicated and improbable symbolic systems as grammar, dictionaries, referential relations,

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logic, the human mind. One of the first tasks for a psychologist, therefore, is to establish some frame of reference within which all these diverse and complex systems can live together in peace and harmony. The engineering approach to the study of communication, which has been so successful in characterizing the transmission of signals, must be replaced, or at least supplemented, by a psycholinguistic approach. I would like first to discuss some of the more general aspects of this new approach, then to consider particular examples of recent psychological research on syntax and semantics that has been conducted under this general conception.

SOME FUNDAMENTAL ASSUMPTIONS

René Descartes in the 17th century formulated modern Western European psychology in terms of a dichotomy between corporeal body and incorporeal soul, thus setting a trap for any subsequent thinker who believed psychology is, or could become, a rigorous science. The usual methods of natural science do not obviously apply to this invisible, intangible, nonextended soul-stuff; it is not necessary to apply them, Cartesians argued, since knowledge of one's soul is given to each man directly by intuition and need not be inferred by inductive logic from scientific experiment.

I mention these somewhat creaky philosophical opinions neither to support nor refute them, but rather because the evidence on which Descartes based his dichotomy provides an appropriate context for the remarks I do want to make.

Descartes said that men differ from animals because only man has a soul; animals do not have souls. Animals and men both have bodies, of course, but bodies are mere machines. An animal differs from man because the animal's body does not interact with an immortal, incorporeal soul. It was the possession of a soul that, for Cartesians, set man off from all other living creatures. Anyone who has grown up in a post-Darwinian world is bound to question this Cartesian conclusion, since evolutionary theory makes man a full-fledged member in good standing of the animal kingdom; there is no biological basis for any sharp dichotomy. Yet the evidence on which Descartes's argument rested was quite clear, and just as valid now as then.

Men have souls and animals do not, according to Descartes, be-

cause men have language and animals do not. As the Cartesians interpreted this fact, language is so subtle, complicated, and influential that it is inconceivable that any mere machine could understand or generate it as humans do. Such an important and uniquely human process as speech could only result from the possession by man of some novel power, some special essence—in short, from a soul.

The evidence Descartes used is still acceptable, although the argument he based it on is not. All men in all societies everywhere in the world speak one or more varieties of human language, and man is the only creature who does so. Other animals have signalling systems, other animals communicate in many interesting ways, but there is something unique about the human animal's communication system that sets it apart from all the others—something that both cloaks and molds the human animal's unprecedented intelligence. In studying language, therefore, we come close to that which is most essentially human about human beings. Nevertheless, there are few modern scientists who would interpret this unique method of communication as an argument for a Cartesian soul. Our possession of language is a consequence of an evolutionary process—an unprecedented but not unnatural event that occurred only in the evolution of man.

The Cartesian argument that language is too complicated for any mere machine must be regarded today as a historical commentary on the conception of a machine that was available to Descartes. Today we know many ways to process natural languages by machine, and the emergence of modern digital computers has enormously expanded our conception of what machines can be and do. True, there is still no machine that deals with language precisely as we do, but at least the possibility of such a machine is no longer inconceivable. So we reject this argument for the Cartesian soul; we have no need for what Gilbert Ryle once called "the ghost in the machine."

Still, there is a fascinating problem here. Descartes was right when he saw that human communication is qualitatively different from animal communication. For psychologists, this difference poses a tantalizing question. How can it be characterized? What have we got that animals lack? Is the difference only quantitative, or is it qualitative? Until we can give clear and satisfactory answers, we cannot claim to understand the human mind. Some progress has been made

toward the answers, I think, but much about our human gift of speech still remains clouded and unclear.

Predication. Generations of philosophers and scientists have struggled with these questions; I will not review their history. In the main, more has been done to rephrase the questions than to answer them.

Perhaps it would be a fair summary of what these struggles have accomplished to say that our human capacity for predication lies close to the heart of the mystery. Grammar begins with the sentence; logic begins with the proposition; both grammar and logic take predication for granted. The problem, of course, is to characterize objectively what such a reformulation could mean. A biologist or psychologist should not take predication for granted; for these sciences predication is a natural phenomenon demanding an explanation.

To speak of predication in particular, rather than of speech in general, may narrow our search, but does not provide our answers. The sad truth is that we are still unable to give definitive solutions for these old and important puzzles. But I want to argue that a scientific formulation of them, in modern psycholinguistic terms, at least suggests how some answers might be found.

Consider this question: when a monkey warns the rest of its troop that a predator is near, how does the vocal cry differ from the English sentence, "I see a hungry lion nearby"?

The human sentence says both more and less than the animal cry. We feel it would make sense to ask whether the human sentence is true or false, whereas it would never occur to us to ask this of a monkey's cry. But is the appropriateness of truth or falsity an important psychological difference?

Perhaps the warning cry should be translated as "run for your lives," which even in English is not something we would weigh for truth or falsity; advisable or inadvisable, perhaps, but not true or false. Yet even this less specific translation seems to distort what actually occurred.

As Grace de Languna pointed out, the interpretation of an animal cry is tied to its context in a way that the interpretation of a sentence is not, or need not be. The context in which a sentence is produced and the context in which it is received can be made completely arbitrary and independent of one another; such freedom

is not available for most animal communication. Perhaps this freedom is the major psychological consequence of having propositional language.

When we equate a monkey's cry to a human sentence we project human psychology in a most imperialistic way. How far do we dare to carry anthropocentrism? Can we go so far as to translate a monkey's silence as "I do *not* see a hungry lion nearby"? The suggestion is absurd; think of all the things that a monkey's silence means it is not seeing! The monkey's cries and silences neither affirm nor deny anything about a lion—they are simply *not* predications. Something essential—and essentially human—is lacking in the cry, but unavoidable in sentence.

If we accept this intuition that there is something more to human speech than to animal cries, we should try to specify what the "something more" consists in. One obvious possibility is that the difference can be attributed entirely to the greater combinatorial productivity of human language. Speech combines elements freely into an unlimited variety of significant sequential patterns. Animal cries are, by and large, relatively stereotyped and invariant, and a sequence of them, unlike a sentence, is little more than a list of vocal responses.

The combinatorial productivity of human language is obviously important, yet in and of itself productivity does not explain the difference between men and animals. *Why* must human signals be more various? What special human need does this combinatorial versatility serve? One answer, in very general terms, is that a highly productive, combinatorial system of signals can free communication from the context of the immediate environment in which it occurs. Human language is characterized by sentences that combine a topic and a comment on that topic; that is what we mean by predication. In its most primitive form, perhaps, the topic or the comment can be supplied by gestures, or by pointing to things nearby. But gestures are tied to the context in which they are produced. In order to gain freedom from the context of communication—to provide vocal substitutes for all possible gestures—a great variety of descriptive signs are needed, enough signs to name every thing or aspect of a thing about which some comment might be made.

Nominalization. Yet even that cannot be the whole story, for human language is far more productive than our freedom from the communication context would lead us to expect. The productive

character of human language is greatly enhanced by the fact that one sentence can become the comment, or the topic for a comment, in a second sentence.

This recursion may sound complicated, but it is not. Consider an example: "Mary sings" is a sentence formed by introducing Mary as a topic for a comment about her vocalization; in "Mary's singing is loud," the sentence "Mary sings" has been nominalized—made into a name in order to serve as the topic of another sentence—and combined with a comment about auditory magnitude. The game can be continued "Mary's singing is loud" can be nominalized and made the topic of "It surprised John," as in the sentence, "The loudness of Mary's singing surprised John." And then "John's surprise at the loudness of Mary's singing was obvious," and so on and on.

"John's surprise at the loudness of Mary's singing" is not a sentence; it is a name, just as "John" and "Mary" are names. By this device a human language acquires infinitely more names than nouns. It is an interesting psychological question to ask whether predications and nominalizations are understood in the same way; whether "Mary sings" and "the singing of Mary" are cognitively different in any way. Combinatorially, however, the point is that predication by itself is not very productive, since it combines topics and comments only two at a time, but taken together with nominalization it becomes infinitely productive.

What logicians generally mean by predication is that a comment is made about a topic—is affirmed or denied of it—in such a manner that any person who understands the predication will recognize the conditions under which it would be true or false. If we were to accept this relation as fundamental for linguistics as well as for logic, then rules of grammar in any particular language would be viewed as machinery whereby: (1) predications were embodied in pronounceable sentences; and (2) predications were nominalized to serve as constituents in more complex predications.

There have been objections to this conception of grammar. For instance, some sentences—imperatives and interrogatives—are not propositions, so grammar would seem to have more machinery than the bare minimum necessary for predication. Yet all these non-propositional sentences are, more or less directly, derivable from predicative origins. "Close the door" is neither true nor false, yet

it is grammatically kin to "You will close the door," which can be true or false. Similarly, "Who closed the door?" is not a proposition, yet it is grammatically related to "Someone closed the door," which is. At a deeper level, therefore, the predicative structure of such sentences is still understood by any person who knows English.

It is this predicative aspect of language that is unique to human communication and not found in other animals. Most of the complex grammatical machinery of human language is entailed by the need to actualize this predicative relation in pronounceable form.

In order to describe this uniquely human mode of communication, therefore, we must deal on the one hand with rules that govern the formation and transformation of predicated relations into sentences, and on the other hand with rules that govern the meaningful interpretation of words and sentences. In philosophical terms, we must deal with syntax and semantics. In linguistic terms, we must deal with grammar and lexicon. These two central aspects, therefore, provide an organization for the psycholinguistic research I want to describe.*

I hope it is obvious that I intend to raise more questions than I answer; do not expect from me any revelation of the ultimate source of the mysterious power language gives. The most I can offer is a new way to ask some old questions. And perhaps I will cause you to think.

SYNTAX

Let me begin with syntax. I would discuss it, not as a philosopher or linguist, but as a psychologist concerned to understand the cognitive processes whereby native speakers of a language conform so accurately and unconsciously to the intricate patterns described by grammarians (psyntax?). In order to give a complete account of our syntactic skills, of course, we would need explicit and detailed

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* A full discussion would treat at least three aspects of language, since phonology could not be ignored in any comprehensive discussion of spoken communication. By avoiding it here I do not wish to give a false impression that all problems of phonetics and phonemics, or articulation and perception of speech, of electroacoustic transduction and transmission of speech waves have been solved. But it is certainly true that those problems are better understood than are the syntactic and semantic aspects which play such a crucial role in the communication of meaning.

information about all of the known grammatical regularities. Even if the necessary grammatical knowledge were at my fingertips (and it is not), this would not be the time or place to display it. However, a bare minimum of grammatical knowledge is necessary if I am to discuss the topic at all. So let me quickly illustrate the sort of grammatical knowledge that all of us, as speakers of English, must share, at least implicitly, if not explicitly.

Constituent structure. Take a simple, declarative sentence, "Bill hit the ball." It contains four words, and their order is important. Rearrange the order of these words and you get something very different, e.g., "The ball hit Bill" is a sentence with a different meaning, and "Ball Bill the hit" is no sentence at all. One thing a grammar of English should tell us, therefore, is why some orderings of words make admissible sentences and other orderings do not.

One approach to this aspect of language is to assume that messages are generated "from left to right," one word at a time, and that each successive word is chosen in the context of the preceding words. This conception of a message source (as a Markov process) has been widely used in information theory, and has many advantages for the statistical analysis of speech. Unfortunately for linguists and psychologists, admissible linguistic patterns are more complicated; a Markov process does not provide a valid characterization of the grammatical or cognitive structure of our sentences.

Consider once again, "Bill hit the ball." The words seem to go together in groups; "the ball" is a natural group and "hit the" is not. Linguists describe such grouping in terms of constituent analysis. For example, "the ball" is a constituent of the sentence; we can replace "the ball" by "it" and still have roughly the same grammatical structure. However, "hit the" cannot be replaced by any single word without completely changing the structure of the sentence, and so "hit the" is not a constituent.

If we proceed in this way, we get the constituent analysis presented in Fig. 1, where constituent substitutions are shown above and their grammatical names are abbreviated below. This simple sentence has two constituents, a noun phrase ("Bill") and a verb phrase ("hit the ball"). The verb phrase likewise has two constituents, a verb ("hit") and a noun phrase ("the ball"). And, finally, the noun phrase has two constituents, an article ("the") and a noun ("ball").

1	Bill	hit	the	ball
2	Bill	hit	it	
3	Bill	acted		

Bill	hit	the T	ball N
	V	NP$_2$	
NP$_1$	VP		

FIGURE 1. Example of the analysis of a sentence into its grammatical constituents.

It should be clear that the constituent structure of a sentence is hierarchical in nature, a fact that is made explicit in Fig. 2, where a tree graph gives an easily visualized summary of the analysis. Moreover, Fig. 2 also indicates how this phrase structure might be characterized as a consequence of the grammatical rules of English. Noam Chomsky, linguist and philosopher at the Massachusetts Institute of Technology, has adapted "rewriting rules" from formal logic in order to show how, by following explicit rules, sentences of the appropriate structure might be derived, just as theorems in logic are derived from a basic axiom by applying rules of deduction. We begin with the axiom S, to which we apply rewriting rule F1 to obtain $NP + VP$; then F3 applied to VP gives us $NP + V + NP$; etc., until eventually the string is rewritten as "Bill + hit + the + ball." Since the grammar does not contain rules for rewriting these symbols, at this point we have produced what is called a "terminal string"—i.e., a sentence. Thus rules F1-7 comprise an example of what Chomsky calls a *generative grammar*.

Several warnings must be issued immediately. Rules F1-7 are at best only a tiny fragment of English grammar; they generate a few other sentences, but nothing like the full range of English. For that we would need an enormously enlarged grammar, including some kinds of rules more powerful than any of those illustrated. Many essential facts of grammar have here been deliberately suppressed in
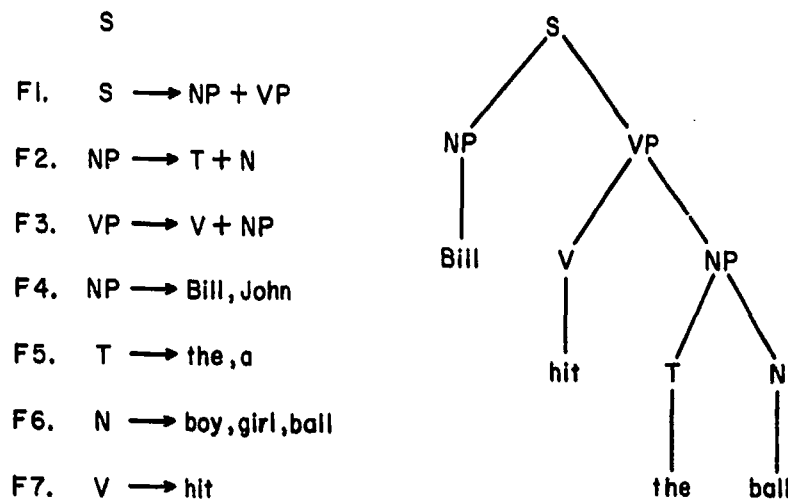
```
      S                                  S
F1.   S  ⟶ NP + VP                      /  \
                                      NP     VP
F2.   NP ⟶ T + N                      |     /  \
                                      |    /    \
F3.   VP ⟶ V + NP                   Bill  V      NP
                                          |      / \
F4.   NP ⟶ Bill, John                     |     /   \
                                        hit    T     N
F5.   T  ⟶ the, a                             |     |
                                              |     |
F6.   N  ⟶ boy, girl, ball                   the   ball

F7.   V  ⟶ hit
```

FIGURE 2. A fragment of the rules for a generative grammar of English, and a tree graph to represent the phrase structure of one of the sentences that the grammar will generate.

order to make the example as intelligible as possible. A fuller treatment of English grammar would have to add extensively to this beginning. In particular, we would need transformational rules; but more about them later.

And—a second warning—the characterization of a generative grammar in terms of a deductive system with rewriting rules, as suggested in Fig. 2, is a formal convenience for the grammarian (and highly suggestive for those who write computer programs), but it may or may not bear any explicit resemblance to what goes on "in our heads" when we produce or interpret sentences. The same structural relations in the sentence can be characterized in several alternative ways, some of which, though less elegant formally, may be more realistic psychologically.

Psychological validity of constituent structure. Regardless of what formal or analytic notation we use to represent it, however, the knowledge that is represented by such rules must somehow be available to people who speak and understand English. Their possession of this knowledge is an empirical fact that can be subjected to test. The psychological validity of constituent structure analysis can be demonstrated in the psychological laboratory in a variety of ways. To illustrate this kind of research, I will describe just one particular

type of experiment; it is but one representative of a variety of demonstrations of constituent effects.

It is a general principle of perceptual organization that irrelevant stimulation tends to interfere minimally with our perception of structural wholes. Garrett provided an auditory example of this principle when he demonstrated that if a string of digits is pronounced with a noticeable pause between some particular pair (thus organizing the string perceptually into two substrings), a short, irrelevant noise imposed on the spoken digits will tend to be reported as occurring during this pause. The irrelevant noise is perceived in such a way as to interfere minimally with the perceptual organization of the spoken digits.

One attractive feature of Garrett's result is that the perceptual displacement of irrelevant clicks can be used as an indicator of subjective organization in instances where acoustic segmentation is less obvious—in grammatical sentences, for example. Fodor and Bever, adapting a technique introduced by Ladefoged and Broadbent, used this indicator to explore the perceptual reality of segments identified linguistically as immediate constituents of a sentence. Their results were consistent with the hypothesis that grammatical constituents are the functional units of speech perception.

Consider the sentence, "That he was happy was evident from the way he smiled." The surface (or constituent) structure is diagrammed in Fig. 3; in the lower half of this figure horizontal lines indicate the extent of the various perceptual units on the hypothesis that these units are grammatical constituents. From Fig. 3 it can be seen that there is a major structural break in this sentence between the words "happy" and "was." The sentence can be (and was) spoken in such a way as to leave no objectively measurable acoustic pause between these two words, but even so, if linguistic structure is a controlling factor, the sentence should be perceived *as if* the speaker had paused at this major constituent break. In that case, extraneous clicks should be judged as displaced toward this perceptual boundary.

Subjects in Fodor and Bever's experiment heard recorded sentences in one ear and a loud click in the opposite ear. The time of the click could be varied, as indicated in Fig. 3, to coincide with the major constituent boundary, to precede it, or to follow it. The listener's task was to write down the sentence (this task forced him to
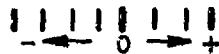
FIGURE 3. Illustrating the click placements used in the experiment by Fodor and Bever.

pay attention to the whole sentence) and then to mark the location of the click. When the results were analyzed, a statistically significant displacement was found in the direction predicted, that is, toward the major syntactic boundary. When the click preceded the major break, there was a tendency to report that it had occurred later than in fact it actually had; conversely, when it occurred following the major break, listeners tended to report that it had occurred earlier than it actually had. Which is consistent, of course, with the hypothesis that grammatical constituents are psychological units and that an interfering click was shifted perceptually so as to interrupt as few perceptual units as possible.

Although efforts were made to avoid it, it is possible, of course, that the perceived groupings reflected acoustic pauses or intonation, rather than the constituent structure assumed by the grammarian. In order to control for this possibility, therefore, Garrett, Bever, and Fodor repeated the experiment with sentences in which exactly the same acoustic stimulus was provided in both cases. The alternative segmentations were suggested, not by physical attribute of the speech signal, but by the context in which the phrase occurred. For example, take a tape recording of the phrase "George drove furi-

ously to the station" and synchronize a click to occur with "George." Now present this stimulus to listeners in the context, "In order to catch his train George drove furiously to the station"; the major constituent boundary is between "train" and "George." As predicted, a click that coincides with "George" now tends to be reported as occurring earlier, at the perceptual boundary. As a control, Garrett, Bever, and Fodor also presented this same acoustic stimulus in the context "The reporters assigned to George drove furiously to the station"; the major constituent boundary now falls between "George" and "drove." Now, as predicted, a click that coincides with George is reported as occurring later, rather than earlier. The click is again shifted toward the constituent boundary, but now in the opposite direction—and *for identical acoustic stimulation*. The perceived boundary must have existed in the mind of the listener, not in the acoustic stimulus.

Such studies as these support the opinion that sentence interpretation is an active process: a listener actively imposes a structural analysis on a sentence, rather than responding more or less passively to some acoustic clues that mark its structure. Moreover, within the limits tested, this active cognitive process seems to confirm the grammatical analysis which was, of course, arrived at on other and very different grounds.

There are other experiments that could be cited to confirm this conclusion, but, since most people are inclined to accept it anyhow, I will not belabor the point that constituent structure has psychological validity. From a psychological point of view, the salient feature of constituent structure is its suitability for expressing predication; its effects on our perception and memory for sentences is only a confirming by-product.

Constituent analysis of sentences into a noun phrase and a verb phrase (as shown in both Figs. 1 and 2) serves directly to represent the cognitive relation between a topic and a comment on that topic, which is the essence of predication. Up to this point, therefore, we can conclude that the linguistic and the psychological characterizations are compatible (even though we may not be entirely certain what the psychological implications are of expressing the grammatical characterization as a rewriting rule). Predication requires a two-part construction, and English grammar provides it handsomely.

Tarsformational rules. However, when we look at more complicated sentences—and nearly every sentence we actually use is more complicated than "Bill hit the ball"—we find it necessary to introduce what Chomsky has called transformational rules.

We have already mentioned nominalization: if we want to characterize the structure of "Bill's hitting of the ball was skillful," the simple way to do so is to exploit the close relation between "Bill hit the ball" and "Bill's hitting of the ball"; indeed, to assume that both the sentence in its declarative form and the nominalization of the sentence are derived from essentially the same underlying structure, but with slightly different transformations applied in the two cases. In a similar way, "The ball was hit by Bill," "Who hit the ball?", "Bill didn't hit the ball," etc., would all be characterized as deriving from the same deep structure, but would have different surface structures because different transformational rules would have been applied in their derivations.

Precisely how transformational grammars should be formulated is a central problem for linguistic theory; one that is currently receiving much attention and about which opinions are developing rapidly. Rather than try to summarize this shifting scene, I would prefer to consider it in terms of the performances that have to be explained, rather than in terms of some more abstract theory about a language user's underlying competence.

In that spirit, therefore, let me describe a psychological interview. The interviewer wears two hand puppets. On his left hand is The Old Woman, on his right, Alligator. He is speaking to a young child.

"The Old Woman and Alligator are going to talk to each other," the interviewer tells the child. "First The Old Woman will say something and then Alligator will answer. You listen closely to what Alligator says, because in a minute you are going to be Alligator and you will have to give the right answers to The Old Woman." The child nods solemnly, and the psychologist makes the puppets talk, as follows:

TOW: Johnny is a good boy.
A: Isn't he?
TOW: His friends are good.
A: Aren't they?

TOW: Tomorrow they will all go on a picnic.
A: Won't they?

The conversation between the two puppets continues in this fashion until the child has heard several instances of *tag questions.** He is then given an opportunity to play Alligator. If he understands that Alligator always responds with a tag question derived mechanically from The Old Woman's sentence, and if he has the grammatical competence needed to make the derivation, a satisfactory performance can be expected from the child. Older children perform perfectly; younger children make mistakes. In this way (among others) Professor Roger Brown and his colleagues at Harvard University have been exploring successive stages in the development of grammatical competence and performance in English-speaking children.

For example, to move directly from The Old Woman's sentence to Alligator's tag response would involve several operations. Take, for example, the pair: "Johnny is a good boy" and "Isn't he?" The operations involved here would be: (1) recognition of the subject of the sentence, in this case, "Johnny"; (2) pronominalization of the subject, which turns "Johnny" into "he"; (3) recognition of the appropriate verb, in this case, "is"; (4) negation of this verb, which turns "is" into "isn't"; and, finally, (5) inversion of the order of

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* It should be obvious that the interview need not be limited to tag questions. There are many types of sentence pairs that can be used in such a test of a child's knowledge. For example, an interviewer can use active and passive voice, with one puppet saying, "Johnny ate the apple," and the other replying, "The apple was eaten by johnny," etc. Or he can use affirmative and negative forms: "Johnny will find find his shoe" *vs.* "Johnny won't find his shoe," etc. Or various other types of questions can be used: "Johnny saw the fight" can be paired with "What did Johnny see?" or "Who saw the fight?" or "Did Johnny see the fight?" Or pairs of sentences from The Old Woman can be combined into one by Alligator: "Johnny came home and Mary came home" can elicit "Johnny and Mary came home," or "Johnny heard the burglar and Johnny called his father" can elicit "Johnny, who heard the burglar, called his father"; or "Mary sings and it sounds pretty" can elicit "Mary's singing sounds pretty," etc., each pair of sentences testing different transformational relations. The list of sentence relations could be extended considerably, so there is no shortage of tasks to set.

The relations between pairs of sentences just illustrated are all grammatical. That is to say, any adequate grammar of English should include an explicit account of the rules for generating all these sentences, and from those rules it should be possible to see precisely what the relations between the sentences are in each instance.

subject and verb, which turns "he isn't" into "isn't he?" Thus we
could imagine the following sequ... ..c:

> Johnny is a good boy.
> Johnny is.
> He is.
> He isn't.
> Isn't he?

Any person who can play Alligator's role and supply the appropriate tag questions must have a working knowledge of these grammatical relations, and of the way they go together to produce the appropriate tag. Recognizing the subject of a sentence may demand a sophisticated analysis; it can be especially tricky when the subject is not explicitly given, as in imperatives: "Close the door" should elicit "Won't you?", even though the appropriate subject and verbal auxiliary, "You will," are both missing from the original sentence.

Correct performance with tag questions also entails considerable ability to analyze verb constructions.

> Johnny will have been running. Won't he?
> Johnny .... has been running. Hasn't he?
> Johnny .... is running. Isn't he?
> Johnny .... ran. Didn't he?

The verbal component used in a tag question is the first auxiliary verb, unless—as in the last case—there isn't any auxiliary, in which case the convenient verb "do" is introduced to play the part of the missing element.

Even negation demands some syntactic sophistication from a successful subject, since the negative must be added when it is missing and removed when it is present:

> Johnny has run. Hasn't he?
> Johnny hasn't run. Has he?

A linguist might be content with a clear and correct description of the formal relations among such sentences, but a psychologist who is interested in how people perform the amazingly skilled acts which underlie even the simplest sentences would like to know more. He

would like to know, for example, what it is that a person does—step by step—in constructing Alligator's response on the basis of The Old Woman's sentence. Does he first recognize the subject, then analyze the verbal construction, then delete, then invert, then pronominalize, then negate? Or does he do it in some different order? Or does he first interpret the proposition underlying The Old Woman's sentence and then use that abstract conception as the input for generating a tag question? Or is it not a serial process at all? These questions are not easily answered. The cognitive processes whereby a tag is added remain stubbornly unclarified, even though their formal consequences are well-known and easily characterized.

One way to make such questions definite and, thus, potentially, answerable, is to phrase them in terms of a computer model. As a psychologist, of course, I have no vested interest in making computers process natural languages, but I do have a real interest in understanding well enough how people do it that I could express that understanding in the rigorous form needed to support computer simulation. There are many people who would like computers to treat language in a humanoid manner; there seem to be many potential benefits to be gained from having such computer programs. My interest in such simulation, however, is merely to test my own understanding of how human beings perform these intricate feats.

Suppose, therefore, that we wanted a computer to play Alligator's role in conversation with The Old Woman. What program of instructions would the computer need? First, we would have to tell the computer how to recognize subjects and predicates in English sentences, which is no easy matter to explain even to students who speak English as their native tongue. When you must explain it to a machine that cannot tell an English sentence from a table of random numbers you must be very precise indeed.

How might we tell a computer to recognize the subject of an English sentence? We might, for example, give the computer a list of nouns, since nouns are often the subjects of our sentences. If "bus" were on the list of nouns, for example, we could tell the computer that when it was the first noun in the sentence, to treat "bus" as the subject and to substitute "it" for "bus" in pronominalizing. This would work for such sentences as "The bus has left," and would give us "The it has left." But wait a moment. What is "The" still doing in the pronominalized sentence? Obviously, "it" must be

substituted for the whole noun phrase, not just for the noun itself. For example, in the sentence, "The first bus has left," we want to substitute "it" for "The first bus," and so obtain "It has left." This is a bit better. But now what do we tell a computer to do about "The bus and the taxi have left"? Should this be converted into "It and the taxi have left"? Probably not. Somehow the computer must be made to understand that English can have compound phrases as subjects of grammatical sentences, and that the proper pronominalization in such cases is "they," not "it." And, of course, we must not overlook the fact that "bus" can serve as an adjective as well as a noun: "The bus driver has left" should not lead to "Hasn't it?"

After an analysis of the kinds of nominal constructions that can serve as subjects of English sentences, it becomes clear that syntactic analysis routines would have to be written for the computer that would enable it to deal with nearly all aspects of English grammar. It seems to be impossible to tell solely from the formal attributes of any word in a sentence, as it is spoken or written, what grammatical function that word may be playing in the sentence. As linguists are fond of reminding us, it is necessary to penetrate beyond the surface structure of the sentence to the deeper relations that underlie it. I will not attempt to develop this argument here, but will merely comment that this penetration beneath the surface structure requires considerable knowledge of English syntax and semantics; enough knowledge, in fact, that we do not yet know how to make all of it sufficiently clear and explicit for a computing machine. Which is a large part of the reason that machines have, thus far, been inadequate to deal with human language the same way human beings do. But we are learning, and as we learn to write better programs, the computers will improve their performance.

SEMANTICS

Now what can be said about semantics? Anyone who aspires to the scientific study of semantics will soon discover that he has almost no theoretical basis from which to begin. Whereas phonological studies of the sound patterns of spoken languages have been well formulated and intensively studied for many years, and linguistic an psychological studies of syntax are currently developing an interesting and respectable body of scientific knowledge, the major

impression you get from a review of research in semantics is of overwhelming diversity and heterogeneity. The problem is not just that we have no theory to build on; there seems to be almost no consensus concerning the range of phenomena for which a theory should be constructed. What we colloquially call "meaning" is no simple, homogeneous thing. Several different problems are confounded together under this heading. In this situation, therefore, it is advisable to begin by narrowing down the subject a bit with some terminological distinctions.

Philosophers generally distinguish two aspects of semantics: *reference* and *meaning*. Reference is concerned with relations between linguistic symbols and other, usually nonlinguistic, entities, states, or processes; a theory of reference must deal with such matters as naming, truth, extension. Meaning, in this context, is concerned with relations among linguistic symbols; a theory of meaning must deal with such matters as significance, synonymity, analyticity, intension. A reader interested in pursuing these topics in a philosophical vein might be well advised to take J. J. Katz's *The Philosophy of Language* as a starting point. There is some disagreement among philosophers as to whether meaning could be reduced to reference if the theories were properly formulated, but I will not try to judge the merits of this argument; here I will respect the usual referential-ideational dichotomy for its didactic value.

Those not familiar with this distinction may find that a simple example, modeled after Frege, will suggest what is involved. Consider the sentence, "Robert McNamara is Secretary of Defense." As of June 1967 the referent of the name "Robert McNamara" and the referent of the name "Secretary of Defense" are identical. In spite of the fact that both names refer to the same person, however, they do not have the same meaning. If they did have the same meaning as well as the same reference, then the sentence "Robert McNamara is Secretary of Defense" and "Robert McNamara is Robert McNamara" would have exactly the same significance. Since they do not have the same significance, it is necessary to distinguish between reference and meaning. This distinction enables us to conclude that two words that refer to the same thing need not have the same meaning.

Now, reference is obviously important for human language, but it is not a unique feature of human language. Many nonlinguistic

stimuli can function as signals by virtue of associations that give them referential character; I have no doubt that such referential associations can be learned by many animals other than *Homo sapiens*. Such associations are, of course, essential for language. However, according to the position taken here, it is not reference, but predication that sets human language apart from the signal systems used by other animals. When we try to establish a semantic basis for predication we are led into problems that belong directly to a theory of meaning, and only indirectly to a theory of reference.

Predication is the affirmation of a comment about a topic. If we take as a central task of semantics to explain how such predicated relations are interpreted, then we are confronted with a problem in the theory of meaning. Presumably, interpretation of a subject-predicate relation must be characterized somehow in terms of the interpretations of its parts and the manner of their combination. That is to say, we would like to characterize the meanings of its constituents in such a manner that, when they are combined, the meaning of the sentence will be projected automatically by a particular rule for combining a subject meaning with a predicate meaning. Perhaps it would not be too misleading to think of this characterization as if it were a problem in mental chemistry; the elements must be characterized in such a way as to enable us to predict which of them can be combined, and what the result of their combinations will be. In order to accomplish this, there must be a structure underlying the lexicon, just as there is an underlying structure behind the chemical table of elements.

The difficulty with this chemical metaphor, from a psychological point of view, is that a limitless variety of presuppositions—facts familiar to and taken for granted by both talker and listener, yet not actually expressed in the sentence—can play a role in understanding the sentence. Only in special cases can the meaning of a sentential compound be specified completely in terms of its semantic elements and their syntactic interrelations; usually information is invoked that has no place in either the lexicon or grammar of English. Speech can be context free, but usually it is not; it is almost never free of shared presuppositions. A better formulation for a psychologist is that a sentence does not contain its speaker's meaning as a sponge contains water; rather it provides some information that a listener can use in constructing a meaning of his

own. If this conception of listening (as a creative process) is accepted, then a complete analysis of sentence meanings into word meanings becomes very difficult, if not impossible.

Whether or not a complete psychological account of sentence interpretation is possible, however, there is a more modest goal that we might hope to attain. Sentence interpretation is not capriciously related to lexical meaning and syntactic function; contexts and presuppositions can be important, but they are not always and everywhere the only determining factors for interpretation. Lexicon and grammar obviously contribute something; their contribution can be isolated from the total psychological process and studied in its own right. Katz and Fodor have suggested that we imagine finding an unmarked envelope containing a sheet on which a single sentence is written, devoid of any indication of source, destination, context, or presupposition. Such a context-free sentence, if it is grammatical and composed of familiar words, will not be completely unintelligible to a person who knows the language. Of course, endless subleties of interpretation may be added when the sentence is put into a context of actual use, but that does not alter the fact that some interpretation can be made even in the absence of contexual knowledge. It is this reduced, but not unimportant process of context-free interpretation that we have in mind when we resort to a chemical metaphor.

Within these limitations, then, a central problem for a theory of meaning is to explain how the meaning of a grammatical compound can be derived from, or characterized in terms of, the meanings of its constituent elements.

One approach to this task is to ask how the meanings of the elements might be characterized. The most familiar answer, of course, is given by lexicographers in the form of a dictionary, where the meanings of words are characterized in terms of explanatory phrases and/or mutually substitutable expressions. Some such answer is a necessary part of any semantic theory of a language, although in our theories we would probably like to make the relations between entries more obvious, and we would certainly like to be more explicit about the rules for interpreting combinations of elements. Our theory, in short, should systematically display and exploit the cognitive structure underlying the lexicon.

What guarantee do we have that the lexicon has structure, or that

any simplification or regularization of it will be possible? The fact that little children must learn the lexicon (and adults remember it) argues that it must be simpler than it looks; no one learns the meanings of words by memorizing dictionary entries as if they were independent and unrelated items in a paired-associates learning experiment. The need for cognitive economy in remembering and thinking argues that there must be some simpler system of concepts and relations underlying the apparent heterogeneity of the dictionary.

Remember where we have come. We asked about the communication of meaning. Our first step was to rephrase this question in terms of the interpretation of predications, and our second step was to ask for an analytic answer, i.e., an account of sentence interpretation expressed in terms of the meanings assigned to the component elements of a predication. The meanings that would be assigned to component elements by a dictionary are necessary but not sufficient for psychological purposes; they make no pretense of representing the cognitive system into which these elements fit. Behind the formal lexicon compiled for purposes of linguistic description there must be a psychological lexicon "in the head" of the language user; this subjective lexical competence bears little if any resemblance to an alphabetic listing of words along with their associated pronunciations and definitions. But what does it resemble? If we tried to imagine what an entry in the psychological lexicon might be like, we would probably propose some kind of triadic constellation that included conceptual, imaginal (perceptual or memory images), and symbolic aspects. Our concept of a frog, our imagery of frogs, and the symbol "frog" are somehow integrated into a psycholexical entity. The psychology of reference concerns the relation between imagery and symbol; the psychology of meaning concerns the relation between concept and symbol. And—most important—each of these complex concept-image-symbol entities is related to, or associated with, many other similar complex entities in some systemic way that we would like to be able to describe. If the imaginal aspects can be set aside, at least temporarily, we might hope for some description of the conceptual relations among symbols.

We would hope to make explicit the psychological structure of the lexicon in such a way that (context-free) meanings of grammatical compounds could be inferred directly from semantic specifi-

cations given for constituent elements and from the manner of their combination. We are still very far from having such a theory for any natural language; most of the discussion so far has been concerned with the general form that such a theory might take if we did have it. Various ways to accomplish the general aim have been proposed and explored in a preliminary fashion.

Theoretical alternatives. Consider a spatial representative of meanings—not because it is correct, but because it offers a frame of reference within which the problem of theory construction can be discussed. In a spatial representation, for example, we might imagine that any particular meaning is a point in some cognitive hyperspace, its position being determined by its values on the set of orthogonal axes defining the space. Then the meaning of a sentence that contains several words might also be a point in the hyperspace computed from the positions of its elements (e.g., their center of gravity). Or the sentence might be represented by some more complicated entity (e.g., a vector, or a directed graph through the component points, etc.) Or, if a metric space seemed inappropriate, we might consider some more discrete kind of "space" having, say, only a finite number of values (usually only two) along each axis; there might be various abstract ways to compound the spaces for individual words into spaces appropriate for phrases and whole sentences. There would be a question as to whether the axes of such a model should themselves be words and/or phrases in the language, or whether it is better to regard the axes as purely abstract conceptions invented by the semanticist for the convenience of his own theory.

Since there is no general agreement about the correct strategy to follow here, it is difficult to discuss the problem intelligibly at such an abstract level; it is difficult to say something substantive without saying more than is justified. Perhaps the best way to give a clearer impression of the conceptual possibilities and difficulties is to mention some examples.

Social anthropologists who have been concerned with this semantic problem have developed something called "componential analysis."* A semantic component is a feature or—to stay with the

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* Different theorists have slightly different interpretations of componential analysis; what is said here will be right in general conception but probably wrong in specific detail.

spatial model—an axis or dimension, usually having a discrete number of values. For example, *sex* might be a semantic component; it would have two values, *male* and *female;* such words as "man," "bull," "son," etc., would all have one value, and such words as "woman," "cow," "daughter,' etc., would all have the other value. The general aim, of course, is to select several such semantic components in such a way that each entry in the lexicon would have its own unique vector of values on the several dimensions, and entries that seemed similar in meaning would share more values in common than would entries whose meanings seemed unrelated.

When they began to work with sets of semantic components, social anthropologists found it necessary to distinguish two different possibilities, the *paradigmatic* and the *taxonomic*. In a paradigmatic system, insofar as possible, every term is given a value on every component. If all the components were binary, this would mean that n components could characterize 2^n different items, which would be a very efficient way to code meanings.

Consider a paradigmatic example. In Fig. 4 a table is given show-

SEMANTIC COMPONENT		Father	Mother	Brother	Sister	Son	Daughter	Uncle	Aunt	Cousin	Nephew	Niece
S:	SEX	0	1	0	1	0	1	0	1		0	1
G:	GENERATION	0	0	1	1	2	2	0	0	1	2	2
L:	LINEALITY	0	0	0	0	0	0	1	1	1	1	1

FIGURE 4. Matrix representation of a paradigmatic semantic system for English kinship terms.

ing how three semantic components might be used to define eleven different terms in the English system of kinship terminology. In Fig. 5 this same classification is given a spatial representation. Anthropologists for various reasons are much interested in kinship—it is related to marriage practices, family and tribal structure, economic relations and religious beliefs, etc.—so this kind of paradigmatic specification is as important as it is economical.

Unfortunately, however, paradigmatic systems seem to be the exception rather than the rule. In most cases a taxonomic structure
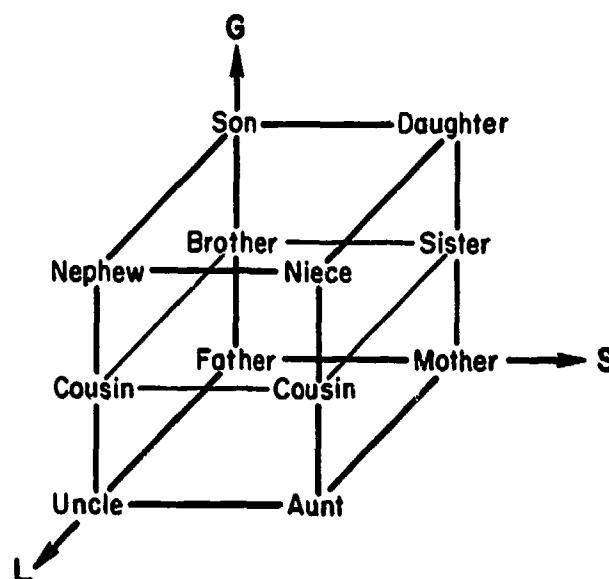
FIGURE 5. Spatial representation of the paradigmatic system given in Fig. 4.

is all that can be established. In a taxonomic system, not every term can be given a value on every component; e.g., if *sex* is to be a semantic component, would "tree" be male or female? Still, there may be a hierarchy among the dimensions of a sort that characteristically leads to taxonomic trees. In order to distinguish paradigmatic dimensions from taxonomic dimensions, let me call the former "semantic components" and the latter "semantic markers."

How a taxonomic tree can result from a system of semantic markers is illustrated in Fig. 6. Here we again have a matrix with items across the top, dimensions down the margin, and cell entries indicating the value of the particular item on the particular dimension. In this case, however, many of the cell entries are blank, which should be interpreted to mean that dimension is simply not relevant to that item. For example, it is simply not relevant to ask whether "fear" should be marked as *living* or *nonliving*, so that cell is left blank. The result of this interaction between dimensions and items is that the dimensions are not used with maximum efficiency: in terms of information theory, semantic markers provide a redundant coding system. The nature of this redundancy is spelled out at the bottom of Fig. 6, where, for instance, it is noted that every item that

SEMANTIC MARKERS

+	−	Choir	Cow	Fear	Mother	Rock	Tiger	Tree	Virtue	...
O: OBJECT	− NONOBJECT	+	+	−	+	+	+	+	−	...
L: LIVING	− NONLIVING	−	+		+	−	+	+		...
P: PLANT	− ANIMAL		−		−		−	+		...
H: HUMAN	− SUBHUMAN		−		+		−			...
F: FERAL	− DOMESTICATED		−				+			...
M: MENTAL	− CHARACTER-OLOGICAL			+				−		...
A: ARTEFACT	− NATURAL	+				−				...
...		:	:	:	:	:	:	:	:	

REDUNDANCY OF MARKING

L ⟹ +O   P ⟹ +L   H ⟹ −P

M ⟹ −O   A ⟹ −L   F ⟹ −H

FIGURE 6. Matrix representation of a taxonomic semantic system for English nouns. The redundancy rules should be interpreted as follows: "Any word that is marked either as living or nonliving is marked + for object," etc.

is marked for *L* (*living* vs. *nonliving*) is also marked + on *O* (is an object): that is to say, if any item is marked for *L*, we know automatically how it will be marked for *O*. Similarly, anything marked for *P* (*plant* vs. *animal*) will be marked + on *L* (will be *living*), etc.

When we examine these redundancy rules, we find that they can be summarized in a tree graph as shown in Fig. 7. For example, the word "tiger" is marked + on *F* (*feral* vs. *domesticated*): the tree graph tells us that "tiger" is also *subhuman, animal, living*, and *object*. The redundancy rules, therefore, can be interpreted as representing what a language user knows about the structure of the lexicon, a kind of basic semantic framework into which new terms can be assimilated as they are learned.

Semantic components and semantic markers, as these terms are generally used, are abstract dimensions: they may have simple and appropriate names, but if so, this fact is irrelevant and unnecessary. Components and markers provide a conceptual framework within which lexical items can be located, much as points are located in a

HIERARCHIC MARKING SYSTEM
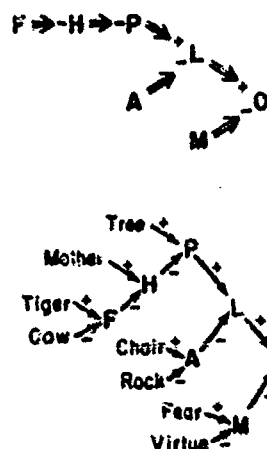
FIGURE 7. The redundancy rules of Fig. 6 are here redrawn to display more clearly their hierarchic structure.

space. There is, however, a slightly different way to think of these relations, a way that may be more agreeable to psychologists, since it seems closer to their traditional concept of association. We can think of them in terms of associations between words. This approach is perhaps one step closer to the usual lexicographic practice of defining one word in terms of other words.

Suppose we were to think of semantic markers as themselves being items in the lexicon, and instead of imagining them to be dimensions in a space, suppose we were to assume a special kind of association that has been learned between them and the words they mark. One very important instance of this special association would be the asymmetric inclusion relation. Under this interpretation we can look at the taxonomic tree in a slightly different way, as indicated in Fig. 8, where all the entries are related by the same kind of inclusion association. Here, for example, there is an association between "tree" and "plant" of the kind we call inclusion, and another inclusion association has been learned between "plant" and "living", etc.

Fig. 8 also represents an associative hierarchy in the form of a list structure. As everyone familiar with contemporary computer programming knows, the matrix and the list are the two basic modes for

FRAGMENT OF AN INCLUSION LIST STRUCTURE
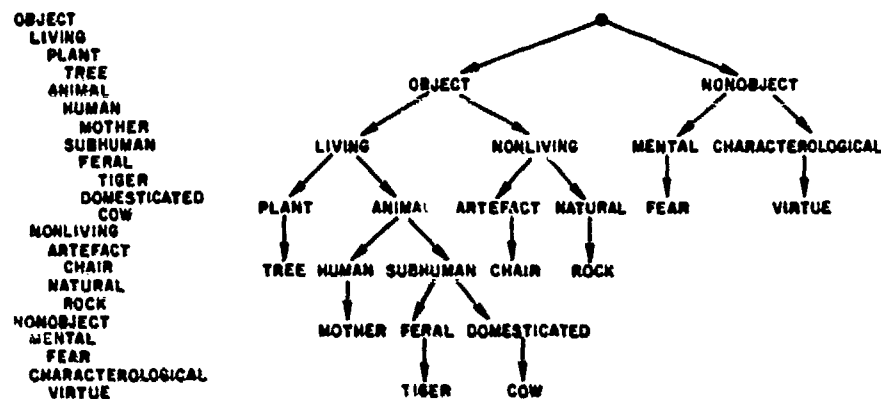
FIGURE 8. The hierarchic taxonomy defined on nouns by an inclusion relation is represented both as a list and as a tree graph. The nodes are here labelled with lexical entries, rather than abstract concepts, as in Fig. 7.

organizing the memory of a computer system. Either matrix or list structures can be used to represent either a semantic marker or a semantic association model of the lexicon. However, I believe that anyone familiar with list-processing languages would be strongly attracted to the list as the appropriate form of organization for this kind of lexical information. List vs. matrix, however, is a tactical question; since we still have important strategic questions unsettled, I do not wish to argue the point.

An associative model, coded as a list structure, however, could also be used to organize the same lexical items in terms of more than one type of association. In addition to the inclusion illustrated in Fig. 8, we might also want to have a part-whole relation of the sort illustrated in Fig. 9. This tree graph (or list structure) represents the fact that a "chin" is a part of a "face," which is a part of a "head," which is part of a "body," which is part of a "person"—relations that must be known to anyone who knows what these words mean. A part-whole association gives us a kind of hierarchic inventory of parts. But note that the strict concept of hierarchy may be sacrificed, as when "neck" is judged to be part of both "head" and "torso"; as long as no loops are permitted, the structure is still weakly hierarchic.

Inclusion relations and part-whole relations are closely tied to

FRAGMENT OF AN INVENTORY LIST STRUCTURE
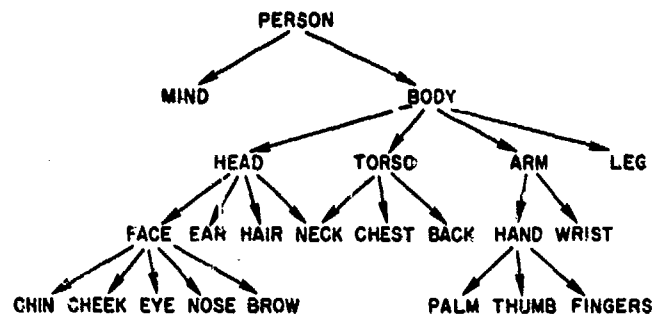
FIGURE 9. A list structure can also be defined by a part-whole relation. Note the presence of "neck" at two places on the list.

important forms of predication. For example, the inclusion of "tree" in "plant" is expressed by the (analytic) proposition "A tree is a plant." If the relation is reversed, as in "A plant is a tree," it is incorrect. Similarly, the part-whole relation between "chin" and "face" is expressed by the (analytic?) proposition that "A face has a chin." Again, if the relation is reversed, as in "A chin has a face," we feel that something odd has been said. This connection of inclusion relations and part-whole relations with particular types of predication is, of course, a familiar fact to those who follow philosophical analyses of language. Its relevance here is that it suggests a way to look at the structure of the lexicon in terms of predication: it is as if the verbs (predicates) imposed a conceptual structure on the nouns (subjects).

These approaches to semantic analysis should give some feeling for the kind of theoretical struggles that are currently going on. The survey is certainly not exhaustive, and undoubtedly not unbiased by my personal interests and research. For example, I have not even mentioned the most famous attempt to summarize the conceptual structure of the lexicon, namely, Roget's *Thesaurus of English Words and Phrases Classified and Arranged as to Facilitate the Expression of Ideas and to Assist in Literary Composition.* Roget has little of theoretical value to offer us. But if Roget was a bit short on theory, he made up for it in energy; he pushed his classification system through the whole lexicon—an impressive and surprisingly useful

enterprise. If we want to improve on Roget, we need not only a better theory, but also a more systematic method of collecting data. Roget presumably relied on his own intuitions about semantic similarity. That may or may not be the best method; it is certainly not the only method.

Empirical methods. Methodology, the bread-and-butter of a scientist working in any given field, is usually spinach to those outside. I will try to keep my methodological remarks as brief as possible.

We now have some notion of what a theory of the interpretation of sentences might look like, and some glimmerings of the kind of lexical information about constituent elements that would be required. It is quite difficult, however, to launch directly into the compilation of a lexicon along the lines suggested by this theory; so many apparently arbitrary decisions are involved that it becomes advisable to try to verify them somehow as we go. One important kind of verification, of course, is given by a theorist's own intuitions as a native speaker of the language; such intuition is probably the ultimate court of appeal in any case. But, if possible, it would be highly desirable to have some more objective method for tapping the intuition of language users, especially if many people could pool their opinions and if information about many different words could be collected and analyzed rapidly. A number of efforts have been made to devise such methods.

As far as I am aware, however, no objective methods for the direct appraisal of semantic *contents* have yet been devised, either by linguists or psychologists. What has been done instead is to investigate the semantic *distances* that are implied by the kind of spatial and semispatial representations we have just reviewed. The hope is that from a measurement of the distances between concepts we can infer something about the coordinates of their universe. Distance can be related to similarity, and judgments of similarity of meaning are relatively easy to get and to analyze. Several methods are available.

The empirical problem is this. It is not difficult to devise ways to estimate semantic similarities among words. On the basis of such judgments of similarity we would like to construct, or at least test, theoretical descriptive schemes of the sort just reviewed. Any large-scale empirical attack on the problem would involve three steps:

(1) Devise an appropriate method to estimate semantic similarities and use it to obtain data from many judges about a large sample of lexical items. These data form a symmetric items-by-items matrix, where each cell a_{ij} is a measure of the semantic similarity between item i and j.

(2) Devise an appropriate method to explore the structure underlying the data matrix. Among the analytic tools already available, factor analysis has been most frequently used, but various alternatives are available. In my research I have used a method of cluster analysis that seems to work rather well, but improvements would be possible if we had a solid theoretical basis for preferring one kind of representation over all others.

(3) Identify the factors or clusters in terms of the concepts in a semantic theory. In most cases, this is merely a matter of finding appropriate names for the factors or clusters. Given the backward state of semantic theory at present, however, this step is almost not worth taking. Eventually, of course, we would hope to have semantic descriptions, in terms of marker or list structures, say, from which we could not only construct sentence interpretations, but could also predict similarity data for any set of lexical items. At present, however, we ha e not reached that stage of sophistication.

Most of the energy that psychologists have invested in this problem so far has gone into step (1), the development of methods to measure semantic similarity. However, since there is no generally accepted method of analysis or established theory against which to validate such measures, it is not easy to see why one method of data collection should be preferred over the others. But, in spite of the sometimes vicious circularity of this situation, I think we are slowly making some headway toward meaningful methods.

There are four general methods that psychologists have used to investigate similarities among semantic atoms: (1) *scaling*, (2) *association*, (3) *substitution*, and (4) *classification*. I myself have worked primarily with classification, but I should mention briefly what alternative procedures are available. Where possible, examples of the methods will be cited, but no attempt at a thorough review is contemplated.

(1) The method of subjective *scaling* known as magnitude estimation, as described by S. S. Stevens in numerous publications, suggests itself as a simple and direct method to obtain a matrix of

semantic similarity scores. So far as I know, this method has not been used in any systematic search for semantic features.

Scaling methods have been used in psychometric research. Mosier used ratings to scale evaluative adjectives along a favorable-unfavorable continuum, and Cliff used them to argue that adverbs of degree act as multipliers for the adjectives they modify. However, these studies did not attempt to construct a general matrix of similarity measures for a large sample of the lexicon, or to discover new semantic features.

One example of the use of scaling is a study reported by Rubenstein and Goodenough. They asked people to rate pairs of nouns for their "similarity of meaning." They used a five-point scale, where zero indicated the lowest degree of synonymy and four the highest. Their averaged results for 65 pairs of words included the following:

cord	smile	0.02
cushion	jewel	0.45
forest	graveyard	1.00
hill	woodland	1.48
magician	oracle	1.82
sage	wizard	2.46
asylum	madhouse	3.04
serf	slave	3.46
midday	noon	3.94

Although Rubenstein and Goodenough did not obtain a complete matrix of all comparisons among the 48 words they used, their results indicate that meaningful estimates can be obtained by this technique.

A difficulty that any procedure must face is that a truly enormous amount of data is required. If a lexicon is to contain, say 10^6 word senses, then the similarity matrix will have 10^{12} cells to be filled. It is obvious immediately that any empirical approach must settle for judgments on strategic groups of word items selected from the total lexicon. But even with that necessary restriction, the problem is difficult. If, for example, we decide to work with 100 items in some particular investigation, there are still 4950 pairs that have to be judged. If we want several judges to do the task, and each judge to replicate his data several times, the magnitude of the data collection

process becomes truly imposing. It is doubtful that judges could maintain their interest in the task for the necessary period of time. If a multidimensional scaling procedure is used in which judges decide which two of three items are most similar, the number of judgments required is even greater: 100 items give 161,700 triplets to be judged.

For this reason, scaling procedures do not seem feasible for any large survey of semantic items; preference must be given to methods that confront a judge with the items one at a time, where similarity is estimated on the basis of the similarities of his responses to the individual items—thus avoiding the data explosion that occurs when he must judge all possible pairs, or all possible triplets. Scaling methods should probably be reserved for those cases where we want a particularly accurate study of a relatively small number of items.

(2) Because of the historical importance of *association* in philosophical theories of psychology, more work on semantic similarity has been done with associative methods than with any other. This work has been reviewed by Creelman and also by Deese, who made it the starting point for a general investigation of what he calls "associative meaning."

In the most familiar form of the associative method, people are asked to say (or write) the first word they think of when they hear (or read) a particular stimulus word. When given to a large group of people, the results can be tabulated in the form of a frequency distribution, starting with the most common response and proceeding down to those idiosyncratic responses given by only a single person. Then the similarity of two stimulus words is estimated by observing the degree to which their response distributions coincide.

The procedure for estimating the degree of similarity from two response distributions is a very general one that has been used in one form or another by many workers. The logic behind it is to express the measure of similarity as a ratio of some measure of the *intersection* to some measure of the *union* of the two distributions. In the case of word associations, the responses to one word constitute one set, the responses to another word another. The intersection of these two sets consists of all responses that are common to the two; the union is generally interpreted to be the maximum number of common responses that could have occurred. The resulting ratio is thus a number between zero and one.

The argument is sufficiently general that intersection-union ratios can be used in many situations other than word association tests; their use in studies of information retrieval, where synonymy must be exploited to retrieve all documents relevant to a given request, has been reviewed by Giuliano and Jones and by Kuhns. The ratio has been invented independently by various workers, for in one version or another it is the natural thing to do when faced with data of this type. Consequently, it has been given different names in different contexts—intersection coefficient, coefficient of association, overlap measure, etc.—and minor details of definition and calculation have occasionally been explored, though rather inconclusively.

The utility of an intersection-union ratio is that estimates of similarity can be obtained without actually presenting all possible pairs to the judges. The assumption underlying it, of course, is that similarity of response reflects similarity of meaning. If, as some psychologists have argued, the meaning of any stimulus is all the responses it evokes, this argument is plausible. But the notion that the meaning of a word is all the other words it makes you think of should not be accepted without some reservations.

The principal recommendation for a word-association technique is its convenience of administration; it is generally given in written form to large groups of subjects simultaneously. The method gives some information about semantic features, since an associated word frequently shares several semantic features of the presented word, but it is also sensitive to syntactic and phonological association. Attempts have been made to classify associates as either syntagmatic or paradigmatic, but the results have been equivocal, e.g., if *storm* elicits *cloud,* or *flower* elicits *garden,* is the response to be attributed to paradigmatic semantic similarity or to a familiar sequential construction? The method is sensitive only to high degrees of similarity in meaning; most pairs of the words elicit no shared responses at all. And no account is taken of the different senses that a word can have; when, for example, *fly* is associated with *bird* and also with *bug,* we suspect that *fly* has been given in different senses by different people, but the data provide no way to separate them.

A variation on the association technique that combines it with the scaling methods has been developed and extensively used by Osgood, who constrains a judge's response to one or the other of two antonymous adjectives. Several pairs of adjectives are used and

people are allowed to scale the strength of their response. By constraining the judge's responses to one of two alternatives Osgood obtains for all his stimulus words distributions of responses that are sufficiently similar that he can correlate them, even for words quite unrelated in meaning. When these correlations are subjected to factor analysis, a three-dimensional space is generally obtained. The position of all the stimulus words can be plotted in a three-dimensional space defined by the antonymous adjectives. It is not clear that these three dimensions bear any simple relation to semantic markers—nor does Osgood claim they should. It is true that words near one another in the space often share certain semantic features, but the method gives little hint as to what those shared features might be. Osgood's method is most useful for analyzing attitudinal factors associated with a word.

(3) Another approach uses *substitution* as the test for semantic similarity. In linguistics a technique has been developed called "distributional analysis." The distributional method has been most highly developed in the work of Zelig Harris.

Consider all the words that can be substituted in a given context, and all the contexts in which a given word can be substituted. A linguist defines the distribution of a word as the list of contexts into which the word can be substituted; the distributional similarity of two words is thus the extent to which they can be substituted into the same contexts. One could equally well consider the distributional similarity of two contexts. Here again an intersection-union ratio of the two sets can provide a useful measure of similarity.

Closely related to distributional similarity is a measure based on co-occurrence. Co-occurrence means that the words appear together in some corpus, where "appear together" may be defined in various ways, e.g., both words occur in the same sentence. We can, if we prefer, think of one word as providing the context for the other, thus making the distributional aspect explicit. A union-intersection measure of similarity can be defined by taking as the intersection the number of times the words actually co-occurred, and as the union the maximum number of times they could have co-occurred, i.e., the number of times the less frequent word was used in the corpus. Co-occurrence measures have the advantage that they can be carried out automatically by a properly programmed computer. Distributional measures can in general be made automatic if a very

large corpus is available—large enough that the two words will recur many times.

Several psychologists have invented or adapted variations on this distributional theme as an empirical method for investigating semantic similarities. It is, of course, the basis for various sentence completion procedures insofar as these are used for semantic analysis. The basic assumption on which the method rests is that words with similar meanings will enjoy the same privileges of occurrence, i.e., will be substitutable in a great variety of contexts.

For example, *couch* and *sofa* can be substituted interchangeably in a great variety of contexts, and they are obviously closely related in meaning. In terms of a theory of semantic markers, some such relation would be expected, since the semantic features of the words in any meaningful sentence are interdependent. The predicate *is upholstered* imposes certain restrictions on the semantic markers of its subject, and only words that have those required features can be substituted as subject without violently altering the acceptability of the sentence. *Couch, sofa, chair,* etc., are substitutable in the context *The . . . . is upholstered,* and are similar in meaning, whereas *sugar, hate, learn, delicate, rapidly,* etc., are not. If the method is used blindly, of course, it can lead to absurd results, e.g., *no* and *elementary* are not similar in meaning just because they can both be substituted into the frame *John has studied . . . . psychology.*

If judges are asked to say whether or not two items are substitutable in a given context, they must be instructed as to what is to remain invariant under the substitution. Various criteria can be applied: grammaticality, truth, plausibility, etc. The results can be very different with different criteria. If meaning is to be preserved, for example, only rather close synonyms will be acceptable, whereas if grammaticality is to be preserved, a large set of words belonging to the same syntactic category will usually be acceptable.

Stefflre has used distributional techniques to obtain measures of semantic similarity. He takes a particular word and asks people to generate a large number of sentences using it. Then he asks them to substitute another word for the original one in each of the sentences they have written. Taking the sentences as contexts and the whole set of substitutions as his set of words to be scaled, he creates a context-by-word matrix, and has his subjects judge whether every context-word pair in the matrix is a plausible sentence. Then

he can apply an intersection-union ratio to either the sets of contexts shared by two words, or to the sets of words admissible in two contexts.

In England Jones has proceeded along different lines toward a similar goal. She uses the Oxford English Dictionary to create a list of synonyms, or near-synonyms, for every word sense, then computes an intersection-union ration for the number of shared synonyms.

A number of workers have resorted to *classification* methods for particular purposes, but until recently there appears to have been no systematic use of these methods to explore semantic features. At present several variations of the general method are in use, but almost nothing of this work had appeared in print at the time this paper was written. In order to illustrate the classification method, therefore, I will describe a version of it that Herbert Rubenstein, Virginia Teller, and I have been using at Harvard.

In our method, the items to be classified are typed on file cards and a judge is asked to sort them into piles on the basis of similarity and meaning. He can form as many classes as he wants, and any number of items can be placed in each class. His classification is then recorded and summarized in a matrix, as indicated in Fig. 10, where data for three judges classifying eight words are given for illustrative purposes. A judge's classification is tabulated in the matrix as if he had considered every pair independently and judged them to be either similar (tabulate 1) or dissimilar (tabulate 0). For example, the first judge, S_1, uses five classes to sort these eight words; he puts "cow" and "tiger" together, "chair" and "rock" together, and "fear" and "virtue" together, but leaves "tree" and "mother" as isolates. In the data matrix, therefore, this judge's data contribute one tally in the cow-tiger cell, one in the chair-rock cell, and one in the fear-virtue cell. The data for the second and third judge are similarly scored, and the number of similarities indicated by their classifications are similarly tabulated in the matrix. Thus, in this example, all three subjects group the inanimate objects "chair" and "rock" together, so "3" appears in that cell; two subjects group the animate organisms "cow" and "mother" together; etc. After the classifications of several judges are pooled in this manner, we obtain a data matrix that can be interpreted as a matrix of measures of semantic similarity: our assumption is that the more similar two

DATA

S_1: (COW, TIGER)(CHAIR, ROCK)(FEAR, VIRTUE)
(TREE)(MOTHER)

S_2: (COW, MOTHER, TIGER, TREE)(CHAIR, ROCK)
(FEAR, VIRTUE)

S_3: (COW, MOTHER, TIGER)(CHAIR, ROCK, TREE)
(FEAR, VIRTUE)

MATRIX

	CHAIR	COW	FEAR	MOTHER	ROCK	TIGER	TREE	VIRTUE
CHAIR					3		1	
COW				2		3	1	
FEAR								3
MOTHER		2				2	1	
ROCK	3						1	
TIGER		3		2			1	
TREE	1	1		1	1	1		
VIRTUE			3					

FIGURE 10. Illustration of how the classifications given by three judges would be tabulated in matrix form for subsequent analysis.

items are, the more often people will agree in classifying them together. In our experience, judges can classify as many as 100 items at a time, and as few as 20 judges will generally suffice to give at least a rough indication of the pattern of similarities.

The data matrix is then analyzed by a procedure described and programmed for a computer by S. C. Johnson of the Bell Telephone Laboratories. The general principle of this cluster analysis is suggested in Fig. 11. If we look at the data matrix of Fig. 10 for those items that all three subjects put together, then we have the five classes shown at the tips of the tree in Fig. 11: (cow, tiger) (mother)

CLUSTER ANALYSIS
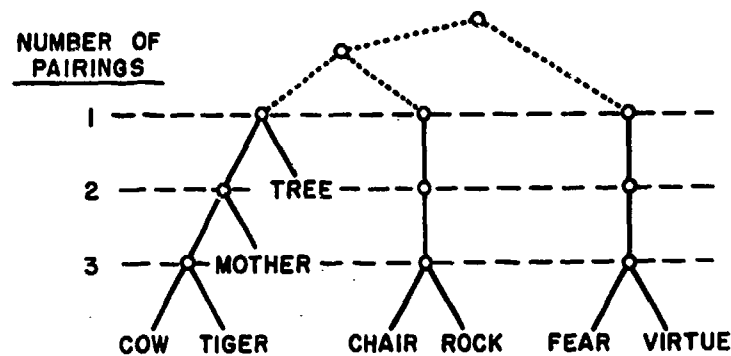
FIGURE 11. Illustration of a cluster analysis of the data tabulated in the matrix of Fig. 10.

(tree) (chair, rock) (fear, virtue). If we relax our definition of a cluster to mean that two or more judges agreed, we have only four classes; "mother" joins with "cow" and "tiger" to form a single class. And when we further relax our definition of a cluster to include the judgment of only one person, we have only three classes: "tree" joins "mother," "cow," and "tiger." As Johnson points out, the level of the node connecting two branches can be interpreted as a measure of their similarity. The dotted lines at the top of Fig. 11 are meant to suggest that an object-nonobject dichotomy might have emerged with more data, but on the basis of the data collected, that must remain conjectural.

Our hope, of course, is that clusters obtained by this routine procedure will bear some resemblance to the kinds of taxonomic structures various theorists have proposed, and that the clusters and their branches can be labelled in such a way as to reflect the semantic markers or dimensions involved. Whether this hope is justified can be decided only by examining the results.

Semantic clusters. In order to illustrate the kind of results obtained with this classification procedure, consider a study whose results are summarized in Fig. 12. Forty-eight nouns were selected rather arbitrarily to cover a broad range of concepts, subject to the constraint that half of them should be names of objects and the other half should be names of nonobjects (concepts). This important semantic

marker was introduced deliberately in order to see whether it would be detected by the clustering procedure; if so important a semantic marker would not come through clearly, then nothing would.
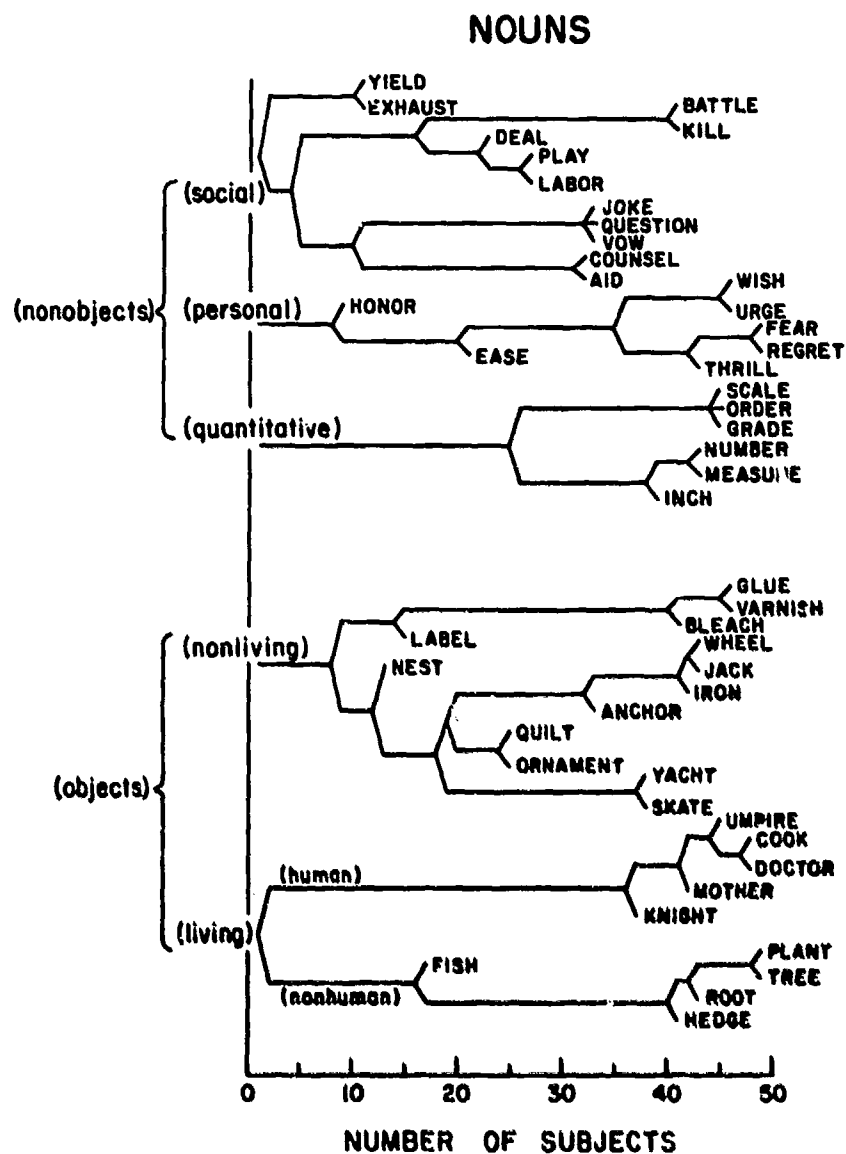
NOUNS

FIGURE 12. Results of a cluster analysis of 48 nouns, with suggested names for the clusters indicated in parentheses.

Each of the 48 nouns was typed on a 3" x 5" card, along with a dictionary definition of the particular sense of the word that was intended and a simple sentence using the word in that sense. The cards were classified by 50 judges, their results were tabulated in a data matrix, and cluster analysis was performed on the data matrix in order to determine what Johnson calls the optimally "compact" set of clusters. The five major clusters that were obtained are shown in Fig. 12, where they are named, quite intuitively, "living objects," and "nonliving objects," "quantitative concepts," "personal concepts," and "social concepts." The finer structure within each of these clusters is also diagrammed in Fig. 12. For example, the tree graph shows that 48 of the 50 judges put "plant" and "tree" into the same class; that 42 or more judges put "plant," "tree," and "root" into the same class; and that 40 or more judges put "plant," "tree," "root," and "hedge" into the same class.

Did the semantic marker that was deliberately introduced into the set of words reappear in the analysis? Yes and no. The clusters obtained did not contradict the hypothesis that our judges were sorting with this semantic distinction in mind, yet their data indicate a finer analysis into at least five, rather than only two clusters, so the object marker is not completely verified by these data. Nonetheless, the results were sufficiently encouraging that we continued to study the method.

The 48 nouns listed in Fig. 12 were also chosen to have the characteristic that each of them could also be used as a verb. In another study, therefore, the verb senses of these words were defined and illustrated on the cards that the judges were asked to classify. When they are thought of as verbs, of course, the object-concept distinction that is so obvious for these words in their noun usages is no longer relevant; the object marker would not be expected to appear in the results of the verb classifications, and in truth it did not. The results of the verb study are not presented here, however, because I do not yet understand them. The object marker did not appear, but neither did anything else that I could recognize. It is my impression that judges were too much influenced by other words in the particular sentences in which the verbs were illustrated. Perhaps the semantic analysis of predicates is basically different from the analysis of subjects: perhaps verbs signify rather special formulae—complex functions into which particular nouns can be substituted as argu-

ments—and the classification of these functions is more complex, more contingent, more difficult than the classification of their arguments. This is a deep question which I am not prepared to discuss here.

There is, of course, an important syntactic basis for classifying English words, i.e., the classification into parts of speech. The data in Fig. 12 were obtained for a single part of speech—nouns—and so do not give us any indication of the relative importance of syntactic categories. Fig. 13, however, shows some results obtained by Jeremy M. Anglin with a set of 36 common words consisting of twelve nouns, twelve verbs, six adjectives, and six adverbs. Twenty judges classified these concepts and the analysis of their data reveals five major clusters. Four of these clusters reflect the syntactic classification, but one—"sadly," "suffer," and "weep"—combines an adverb with two verbs. With this one exception, however, adult judges seem to work by sorting the items on syntactic grounds before sorting them on semantic grounds.

It is important to notice, however, that the results summarized in Fig. 13 were obtained with adult judges. Anglin also gave the same test to 20 subjects in the 3rd and 4th grades, to 20 in the 7th grade, and to 20 more in the 11th grade (average ages about 8.5, 12, and 16 years, respectively). The clusters obtained from the youngest group of judges are shown in Fig. 14. It is obvious that children interpret the task quite differently. When asked to put things together that are similar in meaning, children tend to put together words that might be used in talking about the same thing—which cuts right across the tidy syntactic boundaries so important to adults. Thus all 20 of the children agree in putting the verb "eat" with the noun "apple"; for many of them "air" is "cold"; the "foot" is used to "jump"; you "live" in a "house"; "sugar" is "sweet"; and the cluster of "doctor," "needle," "suffer," "weep," and "sadly" is a small vignette in itself.

These qualitative differences observed in Anglin's study serve to confirm developmental trends previously established on the basis of word association tests with children—an excellent discussion of this work has been given by Doris R. Entwisle—where it is found that children give more word association responses from different syntactic classes than do adults. This trend also appears in the classification data. In Fig. 15 some particular word pairs have been selected
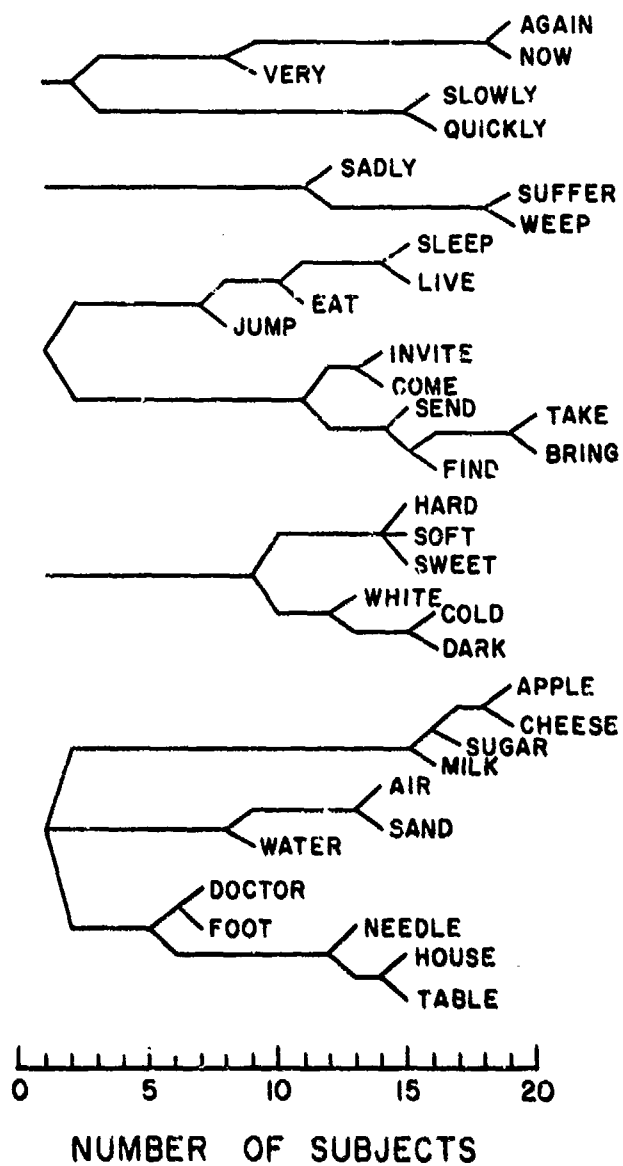
FIGURE 13. Cluster analysis of 36 words by adult subjects. Note that syntactic categories are faithfully respected. Data from J. M. Anglin.

GRADES 3 AND 4
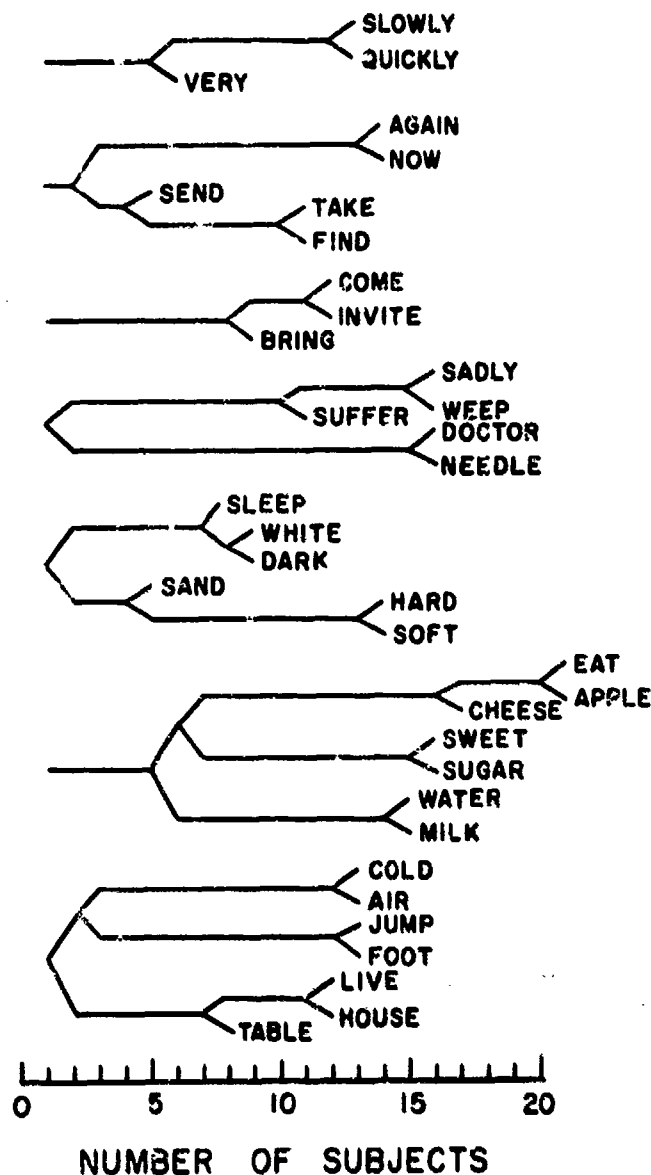
FIGURE 14. The same 36 words of Fig. 13 were classified by children in the 3rd and 4th grades. Note violation of syntactic categories. Data from J. M. Anglin.
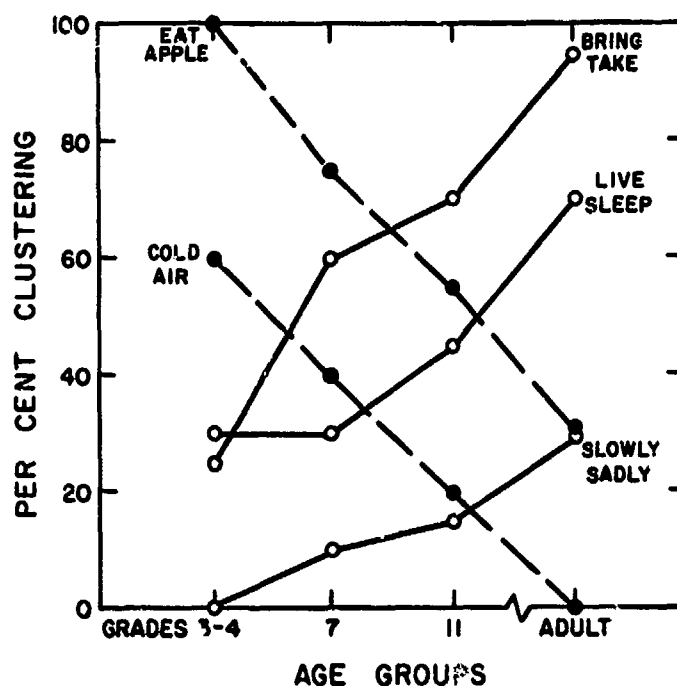
FIGURE 15. Graph illustrating some developmental trends in the classification data. Data from J. M. Anglin.

as illustrating most clearly the changes Anglin observed as a function of age. The thematic combination of words from different parts of speech, which is generally called a "syntagmatic" response, can be seen to decline progressively with age, and the putting together of words in the same syntactic category, generally called a "paradigmatic" response, increases during the same period.

Although there is no basic contradiction between results obtained with word association methods and with word classification methods, some aspects of the subjective lexicon seem to be displayed more clearly with the classification procedures. In order to make some comparison between the two methods, we took word association data collected by Deese and used Johnson's cluster analysis on them. In this particular study, Deese used the word "butterfly" as a stimulus and obtained 18 different word associations from 50 undergraduates at Johns Hopkins. Then he used these 18 responses as stimuli for another group of 50 subjects. He then had response

distributions for 19 closely related words, so that he was able to compute intersection coefficients (a particular version of an inter-section-union ratio) between all of the 171 different pairs that can be formed from these 19 words. Deese published the intersection coefficients in a matrix whose entries could be interpreted as mea-sures of associative similarity between words. When Johnson's clus-ter analysis was carried out on this data matrix, the results showing in Fig. 16 were obtained.

INTERSECTION COEFFICIENTS
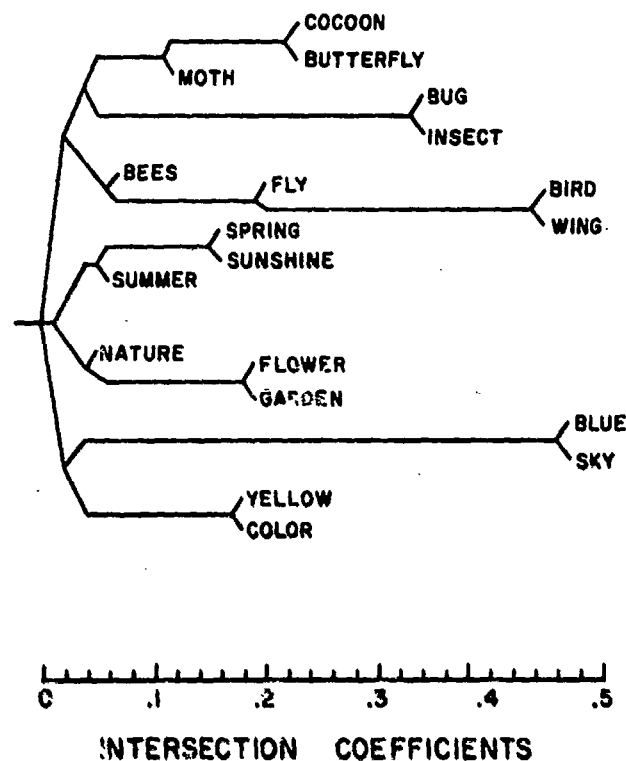
FIGURE 16. The cluster analysis procedure developed by Johnson was ap-plied to Deese's data on the intersection coefficients for word associations.

The clusters obtained with the word association data are a bit difficult to interpret. If we ask whether these clusters preserve syn-tactic classes, the answer depends on whether we consider certain words to be nouns or adjectives. For example, "blue" can be used either as an adjective (as in the phrase "blue sky") or as a noun (as

in the phrase "sky blue"); "flower" would normally be considered as either a noun or a verb, but in "flower garden" it functions as an adjective; "fly" in the cluster with "bird" and "wing" is probably a verb, but it might be a noun; etc. One could also argue that many of the words have multiple meanings; for example, some of the subjects who associated "spring" and "sunshine" might have been thinking of "spring" as a season of the year and others might have meant it as a source of water. In short, data of this sort are useful when words are to be dealt with in isolation, as they often are in verbal learning experiments, but they do not contribute the information we need in order to understand how word meanings work together in the interpretation of sentences.

For purposes of comparison, therefore, we appealed to a lexicographer: the 19 words in Deese's study were looked up in a child's dictionary, where a total of 72 different definitions were found. Each definition was typed onto a separate card, along with the word defined and a sentence illustrating its use. J. M. Anglin and Paul Bogrow tested 20 judges with these 72 items. Their results are shown in Fig. 17. Anglin and Bogrow found nine major clusters, which are quite different from Deese's associative clusters and much closer to the requirements of a semantic theory. For example, there were twelve senses of "spring." Ten of these comprise a single cluster, and the similarity measure suggests how a lexical entry for these ten might be organized. "Spring" in the sense of a source of water did not fit into this cluster, nor did "spring" in the seasonal sense; those two senses would have to have separate entries. Whereas the preceding studies illustrated the use of the classification for widely different concepts, this one indicates that the method might also be useful for investigating the finer details of closely related meanings.

One final example of this method may be of interest. As mentioned previously, Osgood and his coworkers have made extensive use of rating scales defined by antonymous adjectives in order to define a coordinate system in which meanings can be characterized by their spatial position. We decided, therefore, to use antonymous pairs of adjectives in a classification study. One hundred of the adjective pairs Osgood had used were selected and typed on cards—this time without definitions or examples, since the antonymous relations left little room for ambiguity—and 20 judges were asked to

FIGURE 17. Seventy-two senses of the nineteen words studied by Deese (see Fig. 16) were classified and a cluster analysis of the classification data was performed.

classify them. The results for 41 of these 100 pairs are shown in Fig. 18 to illustrate what happened. (Data for the other 59 pairs is analogous, but limitations of space dictate their omission.)

Osgood finds rather consistently that the most important dimension in his semantic differential is the good-bad, or evaluative dimension. Most of the antonymous pairs that were heavily loaded on Osgood's evaluative dimension turned up in our cluster analysis in the three clusters shown in the lower half of Fig. 18. Inspection of these three clusters suggests to me that our judges were distinguishing three different varieties of evaluation which, for lack of better terms might be called moral, intellectual, and esthetic. To the extent that Osgood's method fails to distinguish among these varieties of evaluation, it must be lacking in differential sensitivity.

Fig. 18 also presents a large cluster of adjectives that, according to the introspective reports of some judges, might be considered as going together because "they can all be used to describe people." It is not easy to know what this characterization means, since almost any adjective can be used to describe someone, but perhaps it points in a suggestive direction. It should be noted, however, that this characterization is not given in terms of similarities of meanings among the adjectives, but rather in terms of similarities among the words they can modify. Once again, therefore, we stumble over this notion that the nouns may have a relatively stable semantic character, but the words that go with them, the adjectives and verbs, are much more dependent on context for their classification.

There are still difficulties that must be overcome before the classification method can be generally useful. Some way must be found to work with more than 100 meanings at a time. Some way should be sought to locate generic words at branching points. Effects of context—both of the sentence in which the meaning is exemplified, and also of the context provided by the other words in the set to be classified—must be evaluated. Relations of cluster analysis to factor analysis need to be better understood, and so on and on down a catalogue of chores. But the general impression we have formed after using the classification method is that, while it is certainly not perfect, it seems to offer more promise for semantic theory than any of the other techniques psychologists have used to probe the structure of the subjective lexicon.

NUMBER OF SUBJECTS

FIGURE 18. One hundred pairs of antonymous adjectives taken from Osgood, Suci, and Tannenbaum were sorted by 20 judges; 41 of the pairs are shown here as they clustered under the classification procedure. Note that the good-bad dimension so important in the semantic differential of Osgood is here analyzed into three separate clusters.

SUMMARY

A too short summary of this paper might be that language is what it is because we use it to say things. The capacity to say something--to affirm or deny some comment about some topic—may not be uniquely limited to man, but certainly he is better at it than any other animal. Saying something requires that we have certain grammatical machinery in our languages, ways to combine topics and comments, ways to make one sentence the topic for another comment. Saying something also requires that we have certain semantic machinery, so that what we say can be interpreted by our listener on the basis of what he knows about the meanings of its constituent parts. These are problems that linguists and psychologists share, and that form the kernel of the young science called psycholinguistics. If and when we are able to achieve a deeper understanding of what men do when they say something, we should be able to use that understanding to improve the communication of meaning.

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III. Processes of Interpersonal Accommodation

HAROLD H. KELLEY

THIS PAPER describes some of my research on the processes by which interdependent persons make accommodations to one another. Social interdependence refers to the fact that in most interpersonal relationships, the satisfaction of each person's needs is dependent in some manner upon the actions of the other persons. Social interdependence is, of course, a pervasive characteristic of human life. It ranges from the temporary but severe interdependence a driver endures on the highway in his relations with other motorists, to the equally serious but more permanent interdependence that economic circumstances, laws and social customs, and emotional attachments impose upon married couples. We are interdependent with other persons in our solution of common external problems, in our striving to gain and maintain social status, and in our coping with personal needs and anxieties. This is merely to say that there is ample justification for the careful analysis of social interdependence, if one can ever justify studying a phenomenon by reference to its common occurrence.

Let me first briefly describe my general approach to this research area. The particular type of interdependence that characterizes a relationship is viewed as posing one or more problems for the participants to solve. If they are able to do so, their relationship will be a satisfying and viable one. If not, it will be less rewarding than it might be and may even be so unstable as eventually to

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disrupt. The solutions to interdependence problems consist of those patterns and routines of interaction which insure adequate satisfaction of each participant's needs.

Two very simple examples will serve to illustrate this point of view. Example I, in Figure 1, shows a case where two persons are totally in control of each other's fates. They have mutual fate control.* One convenient way to represent certain aspects of interdependence is by means of a payoff matrix such as is used in game theory. The matrix describes, so to speak, the *problem* confronting
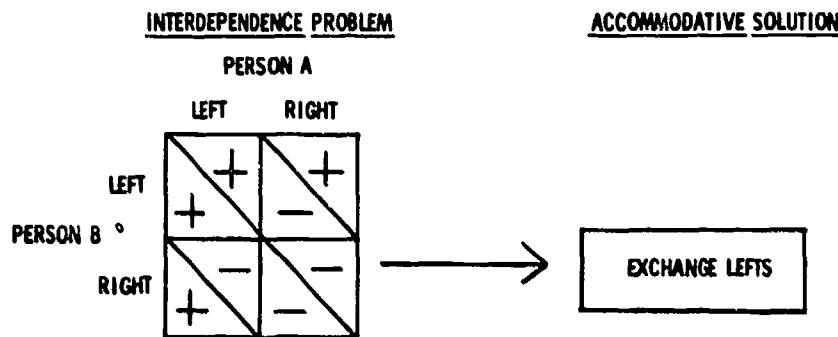
FIGURE 1. Example I Mutual Fate Control.

the participants. In Example I, each of the two persons, A and B, has only two possible responses. The four cells in the body of the matrix show the four possible combinations of their respective actions and the various consequences for each person. The upper right portion of each cell shows the consequences for person A and the lower left portion, the consequences for B. "Plus" means a favorable outcome, "minus," an unfavorable one, and in Figure 2, "zero" means a neutral outcome.

The matrix for Example I shows that each person can provide rewards to the other *without any effect upon his own outcomes,* and that rewards can be provided to both persons at once. This is obviously a simple *interdependence problem* and the indicated *accommodative* solution is equally simple, namely, an exchange or trade of "left" responses. In a real-life instance, "left" might mean paying the other person a compliment, and "right" might mean criticizing

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* The terms for the relationships and much of the analysis here presented is derived from Thibaut and Kelley (1959).

him. A mutual accommodation would require merely exchanging compliments rather than expressing criticisms.

Another simple example is shown in Example II, Figure 2. This matrix represents a case of mutual behavior control which gives the pair *joint* control over their respective outcomes. A relation of this sort might occur when, for some reason, similar responses are incompatible but opposite responses are complementary and rewarding to the person who makes the left response. An important feature of the

INTERDEPENDENCE PROBLEM   ACCOMMODATIVE SOLUTION
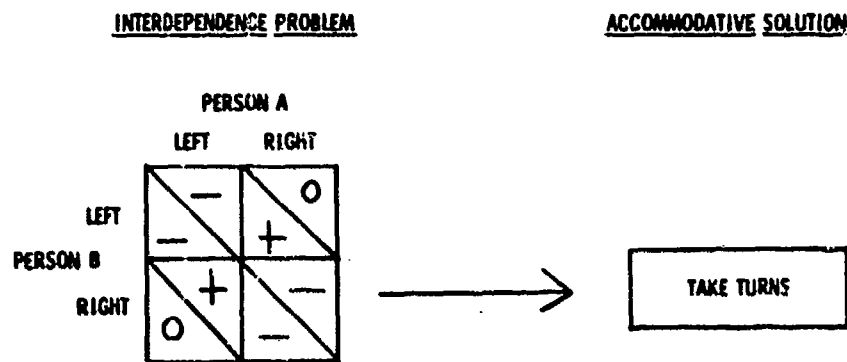
FIGURE 2. Example II Mutual Behavior Control.

relationship is that *only one person at a time* can gain his desired goal. What is indicated as the accommodative solution to this problem is a turn-taking procedure in which the two persons alternate between the left-right and right-left combinations of responses. In a natural situation, "left" might mean being first to use the bathroom in the morning (which enables that person to catch his bus without undue exertion) and "right" might mean using it second with consequent loss of time. Or, "left" might mean listening and learning something from the other person and "right," talking and telling the other something.

These are only two of many possible examples. With slight variations, the mutual behavior control relationship in Example II becomes a pure coordination problem rather than a turn-taking one. In a moment, we'll see a relationship which combines features of these two examples: one person has fate control and the other has behavior control.

One of the relationships that has been subjected to much inves-

tigation in recent years (Rapoport and Chammah, 1965) is known as the Prisoner's Dilemma. This relationship can be formed, as shown in Figure 3, by superimposing upon the mutual fate control pattern of Example I an appropriate degree of control by each person over his own outcomes, which motivates him to make the non-accommodative response. The story that goes with this relationship concerns two prisoners being held as suspected accomplices in a given crime. The district attorney has a weak case against them and they all know it, but he has cleverly placed them in separate cells out of contact with each other. They both know the facts sum-
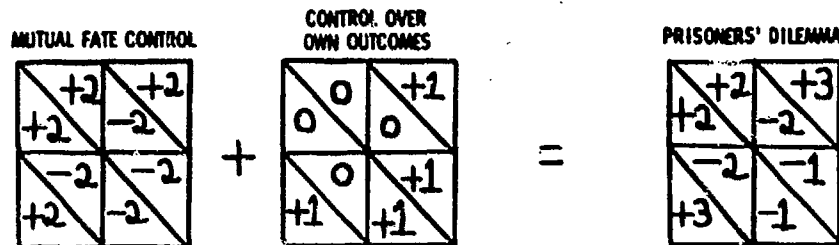
FIGURE 3. Bases of the Prisoner's Dilemma Relationship.

marized in the left matrix; that is, if they maintain silence (left response) they both will go free (indicated by the +2's in the upper left), if one confesses (right), he will be released (+2) and the other one will go to jail (−2), and if both confess, both will go to jail (−2). This mutual fate control relationship would create no problem for the prisoners (they would maintain silence), but the wily district attorney offers each one a special reward (+1 in the next matrix) for confessing; for example, he promises not to press some other charges for which he has managed to unearth evidence. Together these circumstances create the relationship at the right, which is one example of what is known as the Prisoner's Dilemma. The poignancy of this relationship, the dilemma it creates for the prisoners, is that the mutually desirable solution is obvious (in the upper left cell, +2's for both) but is not attainable under these conditions of separate decisions without opportunity for communication. As each prisoner analyzed the logic of the situation in the light of his own interests, he is led to make the "right" or confession choice, even as he realizes that the other person is probably making the same choice and that they are being drawn ineluctably away from the

appropriate accommodation to a solution that is ruinous for both. The Prisoner's Dilemma relationship illustrates one of the many complexities that can be introduced even into the simple two-by-two matrix. Anatol Rapoport, a mathematical behavioral scientist at the University of Michigan, and his colleague Melvin Guyer (1966) have attempted to make a taxonomy of two-by-two games for the simple case where each person's four possible payoffs are simply rank ordered from best to worst. They identify 78 games that are non-identical (no one of the 78 can be derived from another merely by interchanging rows, columns, players, or some combination of these) and they find that the 78 can be classified into 10 categories. In addition to the Prisoner's Dilemma, other types of games in their taxonomy have become identified by such titles as "Chicken" or "Brinksmanship," "Let George do it" or "Hero," "The Battle of the Sexes," and "Inspector-evader." The point of all this is that even with a simple two-by-two paradigm, a great variety of social relationships can be defined.

In addition, of course, any given relationship can be expanded to permit of more responses and graded variations in outcomes as, for example, Pilisuk and Rapoport (1964) have done for the Prisoners' Dilemma game. It is also clear that for some of the interdependence problems we can specify, the accommodative solutions are not as easily identifiable as the simple examples I have given. Problems arise in this respect when the relationship is such that the generally accepted criteria for what constitutes a "good" accommodation come into conflict (e.g., the notion of maximizing joint outcomes may conflict with the concept of equality of outcomes).

In any case, my two simple examples illustrate the general approach to this area of investigation. In each instance, we have a statement of the *interdependence problem* and a statement of the *accommodative solution*. The problem consists of the particular type of interdependence that characterizes the relationship. The solution consists of an interaction pattern or routine that enables the participants to enjoy the rewards and to minimize the costs inherent in their relationship. Given this perspective, the general goals of the research are (1) analysis of the major types of social interdependence and of the interaction routines that provide accommodations to the problems they pose, and (2) (and most im-

portantly) investigation of the processes by which accommodations are attained.

Now, let me outline the research performed within this framework. The three types of experiments I want to describe deal with two types of interdependence problems: (1) common interest problems and (2) conflict of interest problems. The common interest problems have been studied under conditions of minimal information and minimal communication. The conflict of interest problems have been studied (a) under conditions of full information but partial communication, and (b) under conditions of partial information but full communication. The reasons for my interest in these particular information and communication conditions will become clear as we proceed.

First, then, let us consider *common interest problems*. These are relationships characterized by the existence of one or more mutually preferred cells in the interdependence matrix. The first situation we have studied is that shown in our first example, Figure 1, which is the case of mutual fate control. It seems obvious that persons related in this manner can achieve a stable, mutually satisfactory accommodation—that they can agree upon the exchange of "lefts"—*if* they have full information about their relationship and are able to communicate about it, and *if* they are motivated solely as indicated in the matrix. It is less clear, however, whether the appropriate exchange can be worked out *without* the participants' awareness of their interdependence and *without* communication. The original work on the minimal social situation by Sidowski and his colleagues (1956), raised the question as to whether the accommodation can be achieved with minimal information and minimal communication. This is an important question. Inadequate understanding and poor communication are undoubtedly common enough in the real world, that knowledge about how stable arrangements promoting tthe common welfare can evolve despite such handicaps, might prove to be of considerable value.

The experimental setting is schematically shown in Figure 4. Two subjects, A and B, are seated at tables in two separate rooms. They have been brought to the laboratory independently and they have absolutely no knowledge of one another's existence. Each is shown that he can receive points (as indicated on a counter) and he
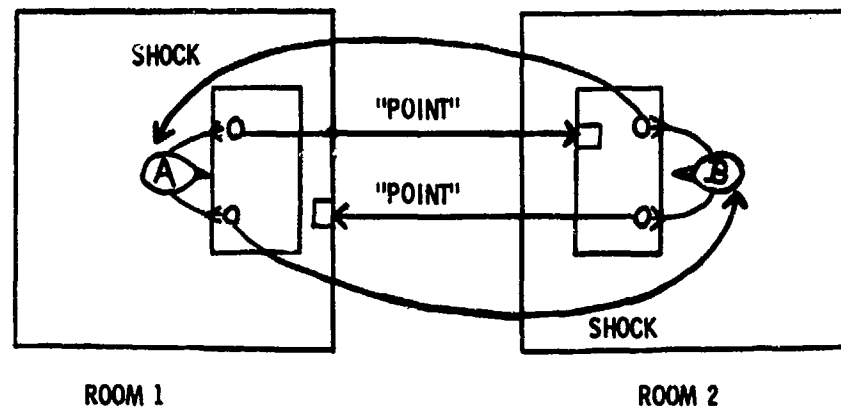
FIGURE 4. Schematic Diagram of Minimal Social Situation.

is told to try to earn as many points as possible. He is also shown that he can receive electric shocks by way of electrodes attached to his arm. One does not need to tell him to try to avoid receiving the shocks. A "Start" signal is given and the subjects begin making their responses. The subjects do not know it, but the buttons are connected as shown in the diagram. Each one's left button gives the other person a point and each one's right gives the other one a shock. The question is, can they learn to give points and to avoid giving shocks without having any other information or opportunities for communication?

The mutual accommodation *can* evolve under these minimal circumstances. Thibaut, Radloff, Mundy and I (1962) were able to show that by following a simple trial and error formula, individuals are able, at least under certain conditions, to solve the mutual fate control problem without either awareness of their relationship or explicit communication. To illustrate, let us assume that when each person makes a response and then receives a positive outcome, he repeats that response and receives a negative outcome, he changes to the other response the next time. In other words, assume he follows the simple reinforcement pattern that has been referred to as a "win-stay, lose-change" strategy. Finally, assume that the two make their responses simultaneously. If one pursues the implications of these assumptions, as in Figure 5, one sees that the sequence of events should follow the pattern of arrows shown there. For example, if on the first trial, A goes left and B goes right, on the next

PERSON A
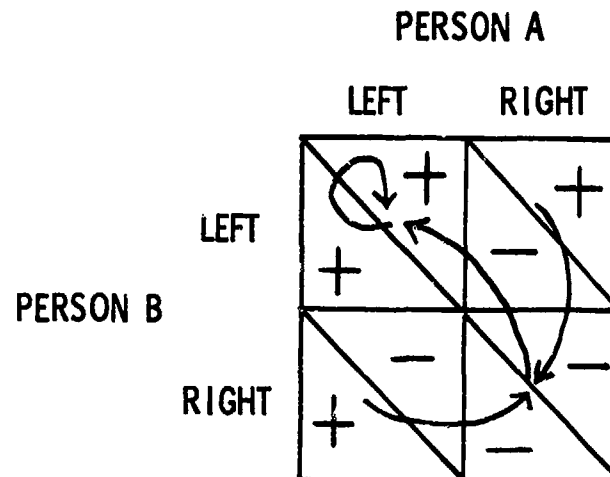
FIGURE 5. Sequence of Events with Simultaneous Responding.

trial, both should go right, the reason being that B, receiving a positive outcome repeats his right response, but A, receiving a negative outcome, changes from left to right. Then on the following trial, inasmuch as both have just received negative outcomes, both should change and go left. When they do so and reach the upper left cell which yields positive consequences for both, they should continue with left responses indefinitely. Our data show that this is essentially what happens. By a simple trial and error process, they are able to learn to accommodate to each other's interests by eventually making mainly left responses. And this is possible without their understanding the relationship and without communication.

In the foregoing, I have assumed that the two persons respond *simultaneously*. Our researches have further shown that the accomodation does not occur if the subjects respond in an *alternating* sequence.

This can be illustrated by Figure 6. Assume, for example, that B has made his left response and A, his right one. Consequently they are in the upper right cell. Inasmuch as B has received a negative outcome, he will change from left to right. Person A now experiences a negative outcome so in turn, he changes from his right to his left. This carries them to the lower left cell. It is A's turn to respond but because he is now receiving a positive outcome, he will persist and repeat his last response which was left. It then be-

PERSON A
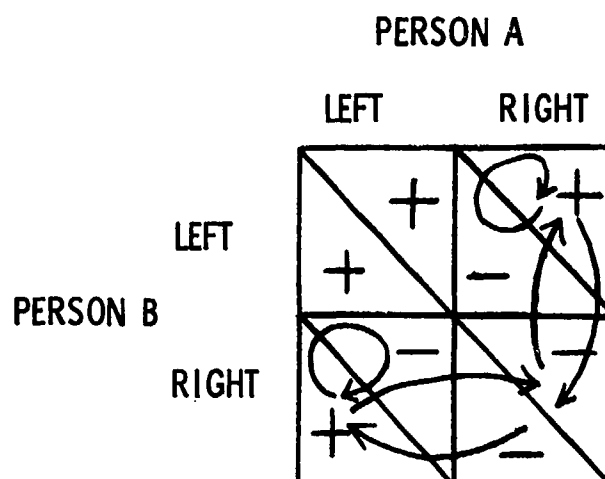
FIGURE 6. Sequence of Events with Alternating Responding.

comes A's turn again and experiencing a negative outcome, he will shift back to his right response. This creates a negative outcome for B so that, when it subsequently becomes his turn, he too changes. And so it continues, cycling back and forth from upper right to lower left, each time passing through the lower right cell. Unless the pair is lucky enough to get in the upper left cell by accident they will not solve the problem. They cannot systematically work their way to that cell. Our experimental data are consistent with this analysis. With an alternation sequence of responding, the accomodative solution is rarely attained for this mutual fate control relationship.

The second interdependence problem we have studied under these minimal social conditions is the mixed fate control and behavior control relationship shown in Figure 7. Person A has fate control over B and B has behavior control over A. I cannot go into the matter in detail but Rabinowitz, Rosenblatt and I (1966) have obtained results from this relationship that are highly consistent with the reasoning I have just outlined. Under conditions of simultaneous responding, accommodation is more difficult to achieve in this relationship than in the case of mutual fate control. (This is something the reader can prove for himself by applying the win-stay, lose-change rule to this matrix.) However, under conditions of *ad lib* responding, when subjects are free to change responses whenever

PERSON A

FIGURE 7. Relationship of Fate Control and Behavior Control.

they wish, the results are reversed. Accommodation is more depend-ably achieved in the fate control-behavior control relationship than in the mutual fate control relationship.

These results are summarized by the table in Figure 8, where I have indicated the quality of accommodation achieved in the two types of relationship (mutual fate control and fate control-behavior control) under three condtions of response evocation (sim·.ltaneous, alternating, and *ad lib*). "Good" means that the accommodation

TIMING CONDITIONS

		SIM	ALT	AD LIB
	MFC	GOOD	POOR	POOR
TYPE OF INTERDEPENDENCE PROBLEM	FC-BC	POOR	?	GOOD

FIGURE 8. Summary of Experimental Results from the Minimal Social Situation.

occurs successfully under minimal social conditions and "poor" means that it does not or only to a slight degree. (The question mark in the FC-BC row under the alternating response condition means that we have no data on that case as yet. We would expect *good* accommodation under those conditions.)

The point I wish to emphasize by reference to this table is the importance of *timing factors* in the achievement of accommodation. We know from a variety of studies that the *type of interdependence, information, communication* and *incentive* are four factors governing the achievement of accommodation. Now we must add a fifth factor to our list, which might be called *process constraints*. These constraints govern the temporal structure of the interaction, the distribution of the process over time, and the order and timing of responses. The process constraints include external constraints (such as those imposed by the experimental procedure or by the calendar or clock) and internal constraints (such as are reflected in natural variations in response times over different situations and different individuals). Our experimental variations of these constraints show that under the minimal social conditions, they interact with the type of interdependence to determine whether or not accommodation will be achieved.

Anecdotes from natural relationships also suggest that timing of actions is often a controlling factor in the course of a relationship. Quarreling lovers must feel like "making up" at the same time; nations lose an opportunity to resolve a dispute because, when one is willing to negotiate, internal political events make it impossible for the other to do so; a management offer that the union could have accepted yesterday becomes unacceptable today. These examples and our experimental results make it clear that the "structure" of time, the distribution of the process over time, is worthy of special analysis in its own right.

The main point of these studies, of course, is that they illustrate the achievement of accommodation by means of very simple processes. The accommodation occurs without the participants' knowledge of their relationship, without communication, and without their being oriented toward cooperation, each being solely concerned with his own interests. I should note here that similar processes have been identified by Seymour Rosenberg (1953) in his work on contaminated feedback and by Christie, Luce and Macy (1952) who

observed what they termed "locally rational" processes in the implicit development of organization in communication networks.

The existence of these processes in the laboratory may have implications for interpersonal accommodations in natural settings. For example, our results with the mutual fate control relationship (Figure 1) have possible implications for such phenomena as the implicit development of collusion between sellers providing a given product to the same population of customers. The movement into the left-left cell corresponds to their both setting high prices for their product. The results from the fate control-behavior control relationship (Figure 7) have possible implications for mutual accommodation in such interdependence relationships as exist between leaders and their followers. The movement into the left-left cell corresponds to a leader's providing wise leadership in exchange for the support of his followers.

However, the existence of these processes in natural settings—whether they are frequent or rare, and where they occur, if at all—is likely to be difficult to ascertain. Because they require neither understanding of the relationship nor explicit communication about it, their presence would not ordinarily be known to the persons involved. Therefore, information about these processes is not likely to be available in subjective or introspective reports about social relationships. Furthermore, their direct identification in the midst of numerous other more complex processes is problematical. In this latter regard, we have found evidence of a simple accommodative process of this sort, co-existing with "higher level" processes, in a much more complex experimental relationship. This process was observed in studies of bargaining in the "bilateral monopoly" relationship, following the research of Siegel and Fouraker (1960). Dietmar Schenitzki's careful analysis of the bargaining process (reported in Kelley, 1964) revealed that, even as the bargainers were explicitly communicating about the negotiation problem, the pattern of their successive offers and concessions enabled them *implicitly* to attain agreements that maximized their joint profits (which is one criterion of successful accommodation). This occurred while the participants pursued their conflicting interests and without their awareness of or explicit communication about this common interest aspect of their relationship. (Schenitzki's work will be described in more detail later in this paper.)

Now, consider the *conflict of interest* problems. In the present context, these consist, not of perfectly competitive relations (such as zero-sum or constant-sum games), but of relationships in which there is some conflict of interest component—some disagreement as to the most desirable accommodation point. This type of relationship is illustrated by the second example (Figure 2) which portrays a relationship in which the two persons gain their respective highest outcomes in different cells of the matrix. This is what has been described as a mixed motive relationship: the parties have a *common interest* in avoiding the mutually negative cells, but a *conflict of interest* over which of the other two cells to settle on.

Once again, it seems reasonable to assume that with full information, full communication and repeated occasions for interaction, persons who are motivated as indicated in the matrix would have no difficulty in achieving an accommodation. Either the logical requirements of the relationship would prevail (and suggest a turn-taking procedure) or, more likely, the appropriate rule or norm, learned as an effective means of accommodation in other similar relationships, would be obvious as a solution to this problem. For this reason, research on this problem, similar to that on common interest problems, has focussed on the achievement of accommodation while under some handicap, either of partial information or of partial communication.

Our work on conflict of interest problems has dealt first with instances of *full* information but *limited* communication. Full information means that both persons are fully aware of the payoff matrix and know each other's outcomes at every point in the interaction. By limited communication is meant that they may use only the actions on the matrix as a means of transmitting information to each other. They do not have the usual means of verbal communication and must rely on tacit communication, that is, communication by actions or moves within the game.

The research on conflict of interest under full information and limited communication has dealt with (1) interdependent avoidance or escape and (2) interdependent approach. In the first, the interdependence arises from the fact that the two persons must use a limited facility to avoid an impending danger, and in the second it arises from the necessity of their using a limited route to reach separate goals.

One important result in both areas of research has been that the accommodation processes are markedly affected by the magnitude of incentives or consequences involved in the relationship. For our study of interdependent escape we began with Alexander Mintz' clever simulation of a "panic" situation (1951). His experimental task required a number of subjects to attempt to withdraw cones from a large bottle within a short period of time. The bottleneck would permit only one person at a time to withdraw his cone and thereby to escape a dangerous flood of water entering the bottle at the bottom. If several persons attempted to withdraw their cones at the same time, they jammed the bottleneck and risked being trapped in the bottle.

In our experiments, we simulated these bottleneck conditions with electric circuitry. Each of seven subjects was placed in a separate cubicle in front of a console such as is shown in Figure 9. At the start of a trial, each subject's push button and indicator light was red, indicating that he was in danger. The subject was told that unless he succeeded in changing the color of his light to green,
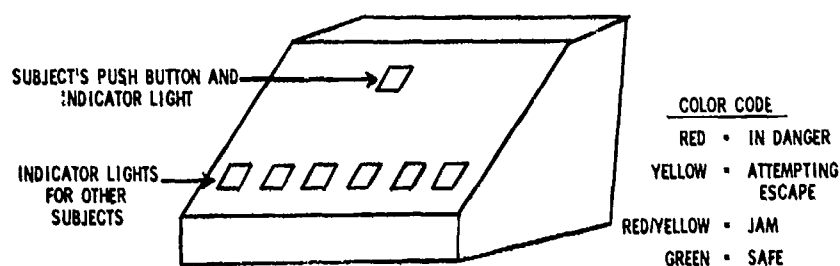
FIGURE 9. Subject Console for the Interdependent Escape Experiment.

before an indefinite time deadline, he would receive some sort of punishment. The subject could attempt to escape from the danger by simply pressing the button, in which case his button turned yellow and an indicator light corresponding to him turned yellow on every other subject's console. If one subject *and one subject alone* held his button down for three seconds, his light turned green indicating that he had succeeded in escaping from the danger. On the other hand if two or more subjects pressed their escape buttons at the same time, the buttons became red and yellow and this indicated a "jam". A jam meant that no one succeeded in escaping

and valuable time was wasted. In short, it was possible for the subjects to escape only by pressing their buttons one at a time and waiting for each person's three-second escape period to elapse. Any simultaneous escape attempts or interruptions of another person's escape attempts simply wasted time without producing any escapes. The seven subjects were given a certain time period in which to escape. They did not know it but this time period was more than adequate for all of them to escape. All who failed to escape before the time ran out were then required to suffer various negative consequences.

Thus, at the start of a trial, each subject was faced with a conflict between waiting for the others to leave and attempting immediately to leave himself. The indicator lights provided him with complete information about what the others were doing but he had no other means of communicating or coordinating with them. This situation obviously requires coordinated behavior similar to turn-taking, specifically, formation of an orderly queue in which each person waits his turn to escape. Thus, the appropriate measure of accommodation is the number of persons in a group who succeed in escaping before the time deadline.

On the basis of his research with very small monetary rewards and fines, Mintz concluded that danger or threat is *not* a necessary factor in the development of jamming and incoordination. Our results indicate that this conclusion is quite misleading. Varying the magnitude of the danger over a fairly sizeable range of negative values, we found that with increasing danger the amount of incoordination increased markedly (Kelley, Condry, Dahlke and Hill, 1965). The results are shown by the data summarized in Figure 10, where percentage of the group escaping is plotted against average amount of concern expressed in the group. This is shown for groups composed of men and women, and under three experimentally induced degrees of threat (low, medium, and high). It is clear that with increasing threat and increasing concern, the amount of jamming increases: fewer persons succeed in escaping before the time limit is reached. It is also clear, by the way, that women are not exactly the preferred accommodation partners under these circumstances. They consistently have the lowest degree of success in escaping.

Mintz' study illustrates that wrong conclusions are easily drawn

FIGURE 10. Results from the Interdependent Escape Experiment.

through the use of insufficiently important incentives. His work also suggests that superficially similar social processes may result from markedly different motivational conditions. Subjects *do* create jams without there being any real danger, but this probably reflects a competition of an aggressive nature (such as is sometimes observed when people queue up in a ticket line or at the exit of a parking lot after the ball game) rather than the fear-based panic observed under extreme danger.

Our research on interdependent *approach* also shows the effects of varying incentives in this type of interdependence relationship. This work has used the Deutsch and Krauss trucking game (1962). As shown in Figure 11, the two subjects in this game find it necessary to share a single path if they are to reach their respective goals in the most economical manner. The two subjects operate two trucking companies, one called Acme and the other, Bolt. In making trips from their respective starting points to their destinations, the two companies have a common, one-way route and each also has a much longer alternate route. Because their costs are a direct function of the time they spend going from the start to the destination, it is in each
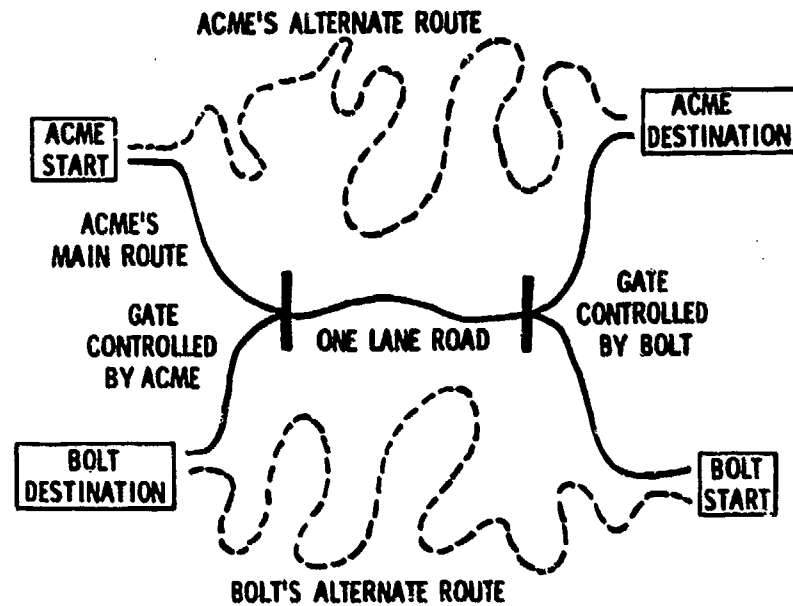
FIGURE 11. Map for the Deutsch and Krauss Trucking Game.

company's interest to use the short, one-way road and to be the first to do so. But, of course, they cannot both proceed by means of the one-way road at the same time. If they do, they block each other's progress and waste valuable time. It is also highly wasteful of time for a company to take its long alternate route. Thus, for maximal profit, one or the other must wait each time while the other uses the short, one-lane road. The structure of their interdependence is again one of mutual behavior control (as in the case of the interdependent escape problem), and accommodation requires coordinated turn-taking. Deutsch and Krauss found that this solution was achieved readily and dependably under ordinary circumstances. However, it did *not* occur when the subjects were both given additional means of interfering with other. These means consisted of the gates (shown in Figure 11) by which a person could block the other person's way to his goal. The detrimental effects of the gates were interpreted by Deutsch and Krauss as indicating that the availability of threat actions operates to interfere with mutual accommodation.

This result was obtained in an experiment where the incentives were quite insignificant, consisting of *small* amounts of *imaginary* money. Gallo (1966) subsequently showed that these detrimental

effects of the gates were greatly attenuated when the rewards obtained upon reaching the goals were greatly increased in value. Figure 12 shows the results. It can be seen that the Deutsch and Krauss results are replicated under the low incentive (imaginary money) condition, the pairs *with* gates doing consistently more poorly than pairs *without* gates. However, when real money is
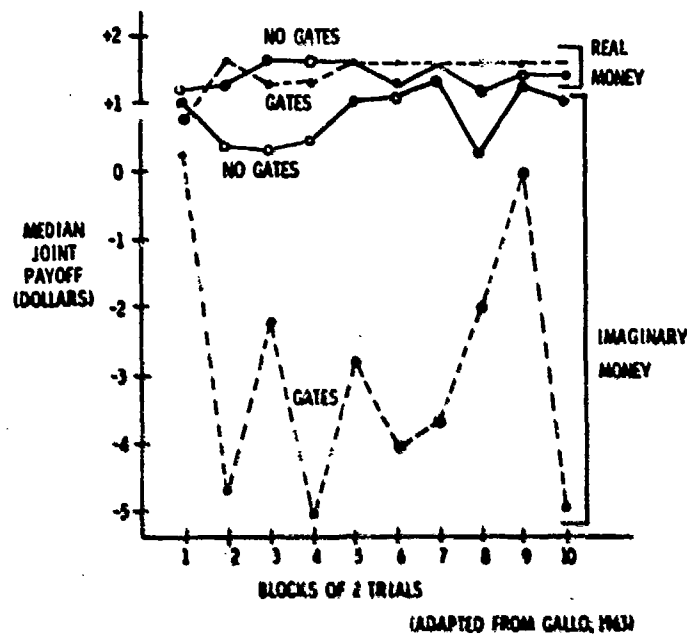
FIGURE 12. Gallo's Results with the Deutsch and Krauss Trucking Game.

involved the gates have only a temporary effect and the two conditions are hardly discriminable in payoffs.

Thus, as in the case of Mintz' work, it appears that conclusions about accommodation drawn from experiments with trivial incentives may be quite misleading. The Deutsch and Krauss conclusion about the detrimental effects of the gates is probably true only when the payoffs are relatively insignificant. It goes without saying that this is hardly the situation about which we are interested in generalizing. As Deutsch has correctly noted in response to this point, Gallo's work shows that if the positive incentives are high enough, persons will suppress or inhibit whatever disruptive motivation or impulses are aroused by the interfering actions. This raises the

whole question, in attempting to generalize such findings, as to the balance between positive and disruptive motivation. This is a question to be answered empirically for any real-life situation. It is not to be taken for granted in advance that the particular balance in the original Deutsch and Krauss experiment, or, for that matter, the different balance in Gallo's experiment, affords the appropriate basis for generalization to any particular situation.

One might be tempted to conclude from our several experiments that high *negative* incentives (such as shock in the escape situation) interfere with accommodation and high *positive* incentives (money in Gallo's work) facilitate it. However, an experiment by Daniels (1967) suggests that this conclusion is not justified. He found that high positive incentives may have *either* facilitative or disruptive effects depending upon other factors in the situation. As his interdependence problem, Daniels used a simple exchange procedure which amounts to a generalized and decomposed form of the Prisoner's Dilemma. Varying the procedural rules and communication conditions, Daniels found that when high value was attached to the items being exchanged, the two persons generally managed to achieve a more profitable mutual accommodation than when the items had little significance. However, high positive incentives tended to have the *opposite* effect when the procedural rules under which the exchange took place made it possible for a person who made an attempt to improve the exchange to be doublecrossed and exploited by his partner.

The point is made more clearly by a subsequent (as yet, unpublished) experiment by Daniels, Meeker, and Shure, conducted at System Development Corporation in Santa Monica. When exchange is terminated by bilateral agreement, as in classical bargaining procedure, the exchange develops more profitably for the participants under high incentives than under low incentives. In contrast, when each exchange can be decided unilaterally, through an action of either party alone, the participants gain smaller profits under high incentive conditions than under low. This suggests the principle that heightened incentives improve the quality of the attained accommodation *only* when the participants have close *mutual* control over the course of the accommodative process.

Our studies of the accommodation of conflicting interests under conditions of full information and limited communication have also

examined tacit communication processes. The experiments with
the situation invented by Deutsch and Krauss have dealt mainly
with the consequences that blocks, gates and other "threat" actions
have for the development of the appropriate coordination routine.
Our research shows that these actions and others that constitute
"threats" in the ordinary sense of the term (communications of
intent to harm the other person) do not necessarily have disruptive
effects upon the accommodation process (Kelley, 1965). In fact, the
effect of such communication possibilities is sometimes to make
possible more rapid accommodation.

Deutsch and Krauss had reasoned that a player could close the
gate to threaten the other person with continued blocking or aggres-
sive action unless he submitted to the player's wishes. However, a
study by Gerald Shure and his colleagues at System Development
Corporation (Shure, Meeker, Moore, and Kelley, manuscript) shows
that a blocking action can communicate other intentions. They
studied an interdependence problem similar to Deutsch and Krauss'
trucking game—one that presents two players with a limited means
of reaching their goals and requires as an accommodative solution
a simple turn-taking procedure. In the absence of other means of
communication, a blocking action (by which one person could pre-
vent the other from using the limited facility) was often interpreted
as a signal given to aid the coordination of their actions. This
interpretation was a common one for Shure's subjects and when the
block was interpreted in this manner, subjects tended to develop
the appropriate turn-taking accommodation to their situation. As
a consequence, pairs who had the block available achieved *better*
outcomes than those without the blocks. In brief, under certain con-
ditions, an action that might seem to be aggressive can be interpreted
as reflecting a cooperative intention. The results of Shure, *et al.,*
are particularly interesting because at the beginning of the inter-
action the subjects tended to view the block in negative terms as a
hostile, selfish, competitive move—much as Deutsch and Krauss had
assumed. However, in many of the dyads, the block action lost what-
ever significance it had initially as a threatening and disruptive
move, and became used regularly as a means of signaling whose turn
it was to use the limited facility.

The present evidence is not entirely clear regarding the conditions
under which the tacit communication significance of an action can

change in this manner. However, there are several suggestive leads in Shure's research and in research reported by Shomer, Davis and Kelley (1966). Shure's results are obtained under conditions that differ from Deutsch and Krauss in one important respect; namely, the subjects had no alternative ways of reaching their goals. If they blocked each other's use of the limited one-way facility, they could not take independent routes to their goals. In other words, they were forced to maintain an interaction with each other. The implication is that when kept in the interdependent relationship and required to persist in their efforts to achieve an accommodation, the block move becomes useful as a means of doing so. Consistent with this view are Shure's additional results from a further experiment. When an alternative, independent facility was provided for each person, the blocks *interfered* with the accommodation just as in the original Deutsch and Krauss experiment. A study by Davis makes the same point, that the interfering effects of the gates depend upon there being available alternative routes which permit independent (though costly) attainment of individual goals. Gallo's results, noted earlier, may be given a similar interpretation. When real money is involved, the subjects are constrained to stay within the relationship and not to take their independent alternative routes. Under these conditions the gates or blocks produce little difficulty. Finally, an experiment by Shomer provides further evidence consistent with these various points: (1) that a threat action is not detrimental to establishment of coordination if interaction is maintained and (2) that under these circumstances the threat action often becomes used as a coordinating signal and facilitates the success of the interaction.

In sum, the indicated generalization seems to be that when other means of communication are not available and when interdependent persons are constrained to the boundaries of their relationship (and are not able to go their separate ways), moves or actions that, under other circumstances, would have negative significance may be transformed into facilitative communication devices.

A further interesting problem in *tacit* communication arose in connection with our work on the minimal social situation which I have described earlier. Although this relationship does not involve a conflict of interest, our study suggests a communication problem that is quite important in conflict settings. In our prior references

to the situation in Figure 4, we were considering the case in which each person had no knowledge of the other in any way. In the study to which reference is now made (also reported in Kelley, Thibaut, Radloff and Mundy, 1962) each person was given a description of the relationship which was complete except for one fact, namely, which of his responses had which effect on the other person. Thus, the two persons were faced with the problem of discovering how they affected one another and once they discovered that, they could easily accommodate to their mutual interests. With a moment's thought, the reader will see how they went about discovering their effects upon each other. They assumed exactly what we have assumed in our analysis of this situation; namely, persons follow a "win-stay, lose-change" tendency. Thus, if a person likes what is happening to him, he will continue with his ongoing behavior; if not, he'll change. To apply this concept for informational purposes, our subject merely pressed one button repeatedly and observed what happened. If the other person's response was stable or consistent (whether he gave a point or shock), our subject could infer that he was making the response that rewarded the other one. If, on the other hand, the other's response was variable, returning now a shock and now a point, our subject could infer that his response was shocking the other one. Once he made this inference, our subject could then use his rewards and punishments to induce the other one to make the proper rewarding response.

However, (and here is where the example illustrates an important point), this method of discovering from the other person's moves how one's own actions affect him can *not* work if both persons are using it at the same time. For example, if both persist in making one response in order to find out what the other one does, each one will conclude he has discovered the good response when in fact he is merely observing the other's similar attempt to gain information. The point is that in order that accurate information be conveyed by this tacit means, it is necessary for one of the persons to have an information-gathering intention to a greater degree than the other one.

The more general principle here is that a person's ability to gain information from the other person's moves depends upon his *own* intentions. Certain pairs of actors' intentions are incompatible in the sense that they *jointly* produce patterns of behavior that are

misleading from the point of view of tacit communication. We are currently investigating within the context of the Prisoner's Dilemma relationship the problem of errors in the inference of intention that occurs as a function of what the two intentions are.

In devoting as much consideration as I have to tacit communication, I do not wish to leave the impression that this sort of communication by actions or moves is highly effective. It is certainly one of the more common findings in this area of research that accommodation is faster and better the more means the interdependent parties have for *explict* communication. This is shown, for example, in a number of Morton Deutsch's experiments (1962) though not, surprisingly enough, with the trucking game (Deutsch and Krauss, 1962). The importance of explicit communication in the interdependent escape situation has been investigated by Arthur Hill (reported in Kelley, Condry, Dahlke, Hill, 1965). Hill's study indicates the value of enabling the less fearful members of a group to express their lack of concern about the danger situation. Specifically, Hill gave the subjects a special response to make if they felt confident and were willing to wait, and this response was incompatible with the escape response itself. The amount of incoordinated "jamming" in the escape route was markedly lower when the less concerned individuals were able to make their presence known, as compared with a situation in which their behavior was not distinctively or dramatically different from that of worried but not yet panicked persons.

Hill's finding raises an important question about tacit communication in situations such as that of the interdependent escape problem, where *imitative* behavior is detrimental to successful accommodation. For example, many danger situations provide a dramatic and obvious way for the most frightened persons to manifest their reactions to the situation. This is the escape response. On the other hand, there is usually no equally dramatic or obvious way for the less frightened persons to exhibit their reactions. The calm persons are often uncertain as to which of several actions to take and rarely do they take the sort of concerted action that the highly fearful persons do. As a consequence, the latter tend to carry the day and to induce the more undecided or neutral persons to follow their example. Hill's research shows the great value that can come from counteracting this situational bias by having available for the more

confident persons a single, obvious, dramatic mode of making themselves known, thereby enabling them to present an example to offset that provided by the concerted escape efforts of the most frightened persons.

The studies just described deal with situations of complete information and limited communication. Consider now the third and last category of research—that on conflict of interest relationships under conditions of *limited* information and *full* communication. The accommodation processes observed under these conditions are usually referred to as bargaining or negotiation processes. The interdependence problems are similar again in general form to Example II (Figure 2) and pose the type of problem for the participants that this relationship would pose if the two persons were not permitted to take turns but had to agree upon a single cell in the matrix for the duration of their relationship. However, our bargaining problems are considerably more complex than this simple 2X2 example, involving more responses and more gradation of outcomes and, therefore, enabling a solution by way of concession and compromise. As noted before, the interdependence problem is "mixed motive" because it is in the two persons' mutual interest to avoid certain states (e.g., the "left-left" and "right-right" combinations in Figure 2), but as between the remaining combinations, their preferences are in conflict.

Most importantly, the accommodation occurs under conditions of *incomplete information*. We have argued that the principal components of the accommodation process observed in bargaining can be traced to this state of incomplete information. This term refers to the fact that although bargainers know the general mixed-motive character of their relationship, they do *not* know directly and at first hand certain important facts about each other's positions and values. Each person knows how much *he* himself values various possible agreements and how poor an agreement *he* can afford to accept without finding it more desirable to turn to other alternative relationships (other customers, employers) available to him. However, he does not know these facts as they pertain to the other person. He can estimate them, inquire about them, and be told about them by his adversary, but in the final analysis all such information is indirect and open to question.

In our bargaining studies, as in natural negotiations, the parties

are permitted to communicate with one another whenever and whatever they wish. However, the conditions just described—the mixed-motive relationship and incomplete information—mean that the participants themselves will place various constraints upon the communication process. These constraints derive from the several dilemmas they face with respect to communication. The *common interest* component of their relationship makes it desirable for them honestly to exchange all the facts about their relationship. Only by doing so can they achieve an accommodation at minimal cost, eliminate the risk of failing to agree when it is in their mutual interest to do so, and avoid agreements that are less satisfactory than others available to them. On the other hand, the *conflict of interest* provides an incentive for each party to conceal or distort information about his own values and position. Thus, it is to each person's advantage to exaggerate how well off he would be if they fail to agree on one of the mutually positive cells and to play down how much he stands to gain if they do so. And as each party feels the temptation to be dishonest himself, he becomes distrustful of communications from the other person.

Another way to put the problem is in terms of *dilemmas* the bargainers face, and to ask how these dilemmas are resolved? With a mixed motive relationship and incomplete information, each bargainer is faced with dilemmas concerning such matters as honesty *vs.* deceit, trust *vs.* distrust, and openness *vs.* secrecy. These dilemmas are paralleled by and intimately related to other dilemmas concerning the proper orientation toward the other party (cooperative *vs.* competitive) and the setting of realistic goals for oneself (high *vs.* low).

The processes by which these dilemmas are resolved have been investigated through analysis of the bargaining behavior of subjects who are dealing with important matters and are relatively free from time pressure in reaching their agreement. This study (Kelley, 1966) was conducted in a classroom setting where students bargained for scores that constituted half of their course grades. They had opportunities thoroughly to master the cognitive aspects of the problem and to rehearse and practice their procedures. And they were given a lengthy time period to reach agreement on a relatively simple contract.

The bargaining task required two persons to negotiate an agree-

ment covering five different issues. An example of the payoff table given one party and from which he bargained is shown in Figure 13. The five different issues (items) are listed across the top of the table and the 20 ways of settling each issue are listed down the side of the table. The table indicates for our player the value to him of an agreement at each possible point. The two bargainers' interests

ITEMS

	(1)	(2)	(3)	(4)	(5)
A	0	101	11	20	21
B	3	98	14	23	21
C	5	95	17	25	21
D	6	92	20	26	21
E	7	88	22	27	21
F	7	83	24	27	21
G	8	78	26	28	21
H	8	71	27	28	20
I	9	62	28	29	20
J	9	48	29	29	20
K	11	39	33	31	20
L	13	36	36	33	20
M	15	33	39	35	20
N	16	29	42	36	20
O	17	25	44	37	19
P	18	20	46	38	19
Q	19	15	48	39	19
R	19	9	49	39	19
S	20	0	50	40	19
T	20	-19	50	40	19

FIGURE 13. Payoff Table Given One Bargainer.

are in direct opposition on each issue because the payoff table for the other bargainer runs in the opposite direction on each item. However, it can be seen that not all issues are equally important to our bargainer. For example, while it is terribly important to our bargainer where issue #2 is settled, he is relatively indifferent about where issue #5 is settled. The relative importance of the five items differs between the two bargainers and it is this feature that converts the relationship from a zero-sum or constant-sum game to the type of mixed-motive game we are considering here. The bargaining task was considered completed only if the two bargainers had reached agreement on all five items before a time deadline. During this period they proposed different contracts or sets of agreement

points to one another and reacted with counterproposals, conces-
sions, and so on. In addition, each bargainer was assigned a total
score he would receive if they failed to reach agreement before the
time deadline or if, as was permitted, either party unilaterally broke
off negotiations. For the payoff table in Figure 13, the score might be
145. These scores provided each person with a minimum guaranteed
level which he was certain to get regardless of the other person's
actions. A final essential feature of the situation was that informa-
tion defining a given party's payoffs and his guaranteed level was
given only to that person, and the bargainers were not permitted to
exchange these official payoff sheets. During the negotiation, they
were free to say anything they wished about their values and to use
any sort of appeal they wished. However, they had no completely
convincing way of communicating their respective values and in-
terests, and thus were faced with the problems of persuasion and
trust.

This bargaining task was given to advanced undergraduate stu-
dents. It was made important to them by making part of a course
grade depend upon their success in a series of such negotiations.
They had ample opportunity to study and think about the stua-
tion, and at each session, they had a 75 minute period merely to
reach agreement on these five items.

The results obtained under these rather ideal conditions indicate
that the bargainer's behavior regularly exhibits certain features
which can readily be interpreted as having functional value in en-
abling the bargainers to resolve the dilemmas inherent in their
relationship. That is, the negotiating behavior we observe seems to
provide means of deciding on an acceptable goal level, a means of
gaining convincing information from the other party without tak-
ing his statements at face value, a means of convincing him of your
own situation without being totally honest and open, and a means
of avoiding agreement on contracts where there exist better ones for
both parties.

These behaviors seem to fall into three categories: (1) avoiding
early commitment, (2) inducing the other to make concessions, and
(3) making economical concessions.

The *first category, avoiding early commitment,* is reflected in four
behaviors: (a) starting high, (b) making exploratory rather than firm
offers, (c) settling the entire contract as a package rather than on an

item by item basis, and (d) using all the time available (bargaining right up to the last minute). These actions have in common the effect of keeping things open and flexible for as long as possible in order to have maximal opportunity to gain information. In view of his uncertainty about the situation and his need for information in order to set optimal goals, a bargainer should not become committed to any aspect of the contract until he has had all possible opportunity to obtain such information. Because the other person can be expected to withhold or distort the relevant information our negotiator can anticipate gaining it only through the course of the interchange, from the patterning and timing of the other's actions rather than from his explicit statements. Thus, the resolution of both the dilemma of goals and the dilemma of trust requires the avoidance of early commitment. The flexibility and tentativeness implied by these commitment avoiding actions is also useful in resolving the dilemma of honesty versus dishonesty. As long as one's own proposals are not made in rigid or final form, they may contain some exaggeration and distortion without running the risk of disconfirming them through one's subsequent actions and thereby, of reducing the other person's willingness finally to believe you.

The *second* general *category* of behavior, regularly observed in our experiment, consisted of *inducing the other party to make concessions*. This involved the application of pressure through such specific tactics as (a) making occasional negative concessions or retreats in one's own offers, (b) using time pressure, and (c) persistent registering of complaints that the other's offers are inadequate. The concessions the opponent makes seem to be essential to our bargainer's resolution of the dilemmas of trust and goals. Each concession heightens his feeling that his emerging share of the contract is an appropriate one. And the increasing grudgingness with which concessions are granted undoubtedly makes increasingly credible the other party's communications about his unwillingness to go further.

Of course, making concessions is a two-way affair. A basic way of eliciting a concession is to make one. Thus, throughout our data from this study and from similar bargaining studies there is overwhelming evidence of a turn-taking or alternation pattern between the two bargainers in their making of concessions. But in making their concessions, our subjects regularly strived to behave according to our *third category*, that is, to *make economical concessions*. An

economical concession means one that gives the other party as much as possible but gives up as little as possible for one's self. Economical concessions are probably important in resolving the dilemma of cooperation. In the attempt to make the other person satisfied while sacrificing as little as possible of one's own values, account is taken of both parties' interests or, at least, of their feelings. This amounts to adopting a quasi-cooperative orientation (with what is often a cooperative effect, as we will see in a moment) without resorting to open and explicit cooperation.

Making economical concessions appears in such specific tactics as (a) seeking information about the opponent's values and priorities (necessary if the optimal concessions are to be made on an informed basis) and in (b) trying out various contracts at a given value level for one's self (which, as we shall see in a moment, makes possible optimal concessions on a trial and error basis). Also, because economical concessions, if perceived as such, are not likely to be highly regarded by the opponent, our bargainers consistently (c) exaggerated the cost to themselves of concessions they made.

The attempt to find ways to make concessions that are most valuable to the other party, but are least costly to one's self has valuable consequences for the joint welfare. This attempt generates a tactic that we had earlier identified in Schenitzki's analysis of the Siegal and Fouraker bargaining situation—that of testing several different contracts at a given level, to see whether any are acceptable to the other person, before making further concessions. This procedure serves to reduce the likelihood that unnecessarily poor contracts will be selected, although the bargainers are not particularly aware of this consequence of their tactics.

This process can be illustrated by reference to Figure 14 which is based on Schenitzki's work and adapted from Kelley (1964). The profits of one player (who was called the "seller" in Schenitzik's study) are shown along the ordinate, and the profits of the other player (the "buyer") are shown along the abscissa. Plotted on the figure are the various contracts the pair might agree upon which are profitable for both (that is, above their respective break-off or independent values). The seller seeks to attain agreement on a contract at the upper left of the scatter plot and the buyer seeks to attain agreement at the lower right. The question that concerns us here is whether, in the pursuit of these interests, they will manage

TIME 1   TIME 2   TIME 3
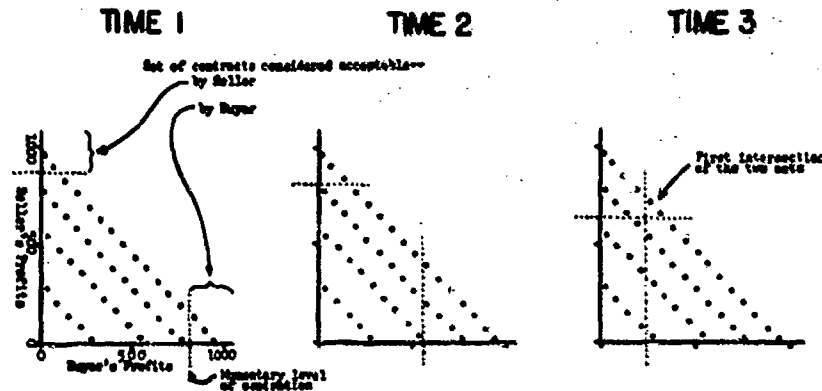
FIGURE 14. Process by which Maximum Joint Outcomes are Attained in Siegel and Fouraker Bargaining Situation.

to agree on a contract yielding them maximum total profit (one on the outer line of contracts, usually referred to as the "Pareto-optimal" outcomes) or one that is less than optimal for the pair (one of the "inner" contracts). The answer would obviously be the former *if* both parties knew both sets of values, but remember that we are considering the case of partial information where each player knows only his own profits for the various contracts. Schenitzki's work shows that despite this limitation of information, if the negotiators start high and make their concessions in a systematic manner, they will agree on a contract characterized by Pareto-optimality. This can be seen in Figure 14 as follows. Knowing only the profit each contract yields him, each bargainer has little choice but to begin by proposing contracts for which his own profits are high. As he finds these to be unacceptable, he proposes additional contracts yielding him somewhat lower profits and so on. As he makes successive concessions, each time dropping his level of aspiration, he enlarges the set of contracts he considers acceptable. Thus, as in Figure 14, it can be seen that as time progresses, the set of contracts the buyer considers acceptable becomes larger, and the same is true for the seller. The question of predicting their final agreement resolves into the question of where these two sets of acceptable contracts will first overlap. As shown in Figure 14, the two sets are likely to intersect first on the contracts located along the outer edge of the total set. Thus, if concessions are made systematically, with fairly good testing of the various possible contracts at each profit

level before moving on to less profitable ones, the common interest will be served to maximal degree. It should be noted that this process of systematic concessions identified by Schenitzki is possible in the bargaining situation we have been considering only if the negotiation is conducted *in terms of packages* or sets of items rather than one item at a time. If the subjects formally settle one or two of the various issues before dealing with other issues, they lose the flexibility that is necessary for systematic exploration of each profit level. They thereby reduce the likelihood that they will agree upon the optimal contracts.

The preceding observations refer to highly motivated and skillful bargainers, working under relatively little time pressure. In the absence of these rather ideal conditions, we might expect that the restraints against exchange of information would not be adequately circumvented because there would not be the kinds of extended testing, probing, and exploring that has just been described. If bargainers are unable to exchange information about their relationships (because their less than maximal efforts to do so are thwarted by their tendencies to withhold information), how do they solve their accommodation problem?

A recently completed series of studies (Kelley, Beckman and Fischer, 1967) seems to shed some light on this question. Two parties negotiated the division of a reward, again under conditions of incomplete information. For each one, there was specified a minimum share of the reward which it was necessary for him to obtain if he was to make any profit on the agreement. Each knew his own minimum necessary share, but not that of his adversary. Each pair bargained on repeated problems with the two minimum necessary shares varying unpredictably from one occasion to the next. The stakes were not such as to warrant extended bargaining, and in any event, the time pressure was high enough to preclude extended bargaining.

These studies yield highly regular patterns of results in terms of such things as opening bids, order of successive bids, rates of concessions, time to reach agreement, points of agreement, and failures of agreement. What is particularly striking about the results is the degree to which they are reproduceable by a simple mathematical model. The details would not be appropriate here, but the properties of this model suggest that, in the absence of information about

the situation of his opponent, the bargainer's behavior is based upon general expectations he forms about the relationship. These expectations are geared to the situation in general and to the typical adversary. As such, these expectations afford generally reasonable guide lines for making accommodative concessions while still protecting one's interests. Their only limitation is that, being adjusted to the average or typical situation, they are not appropriate for atypical cases. The consequence is that appropriate adjustments are not made for these unusual cases. To make such adjustments would require information exchange, and little such exchange occurs under the conditions that prevailed in this experiment. The consequent cost to the pair is a high probability of failure to reach accommodations in certain situations where agreement is in their mutual interest (although only marginally so). In this respect, these experiments particularly highlight the difficulties negotiators encounter in achieving satisfactory accommodations when their respective expectations as to shares of the reward are both high. With open and dependable communication, the expectations would be adjusted to the two sets of needs (the minimum necessary shares) that the bargainers bring to their relationship, and the difficulties arising from too high demands cou'd be avoided. In the absence of open and dependable communication, the bargainers can evolve methods that enable them generally to make accommodations, *but* these methods yield failures of accommodation when their circumstances depart too sharply from the average or typical.

SUMMARY

This wide ranging set of experiments merely samples the total domain of phenomena that eventually must be explored in order to fulfill the goals stated at the outset, namely, (1) to identify the types of interdependence problems and their accommodative solutions, and (2) to analyze the processes by which accommodations are made.

The four or five main categories of factors to be studied and analyzed in their interplay have become fairly clear from the work so far. These are (1) pattern of interdependence, (2) information held by the participants, (3) their communication capabilities, (4) the incentives and motivation, and (5) what have been called the

process constraints: the factors governing the evocation of responses as to their order and timing.

The research described here is part of the new trend of experimental work oriented toward developing a basic science relevant to interpersonal and intergroup relations. The special contribution of social psychologists to this field has been the development and application of appropriate experimental methods. However, it should be emphasized that the experimental work does not proceed in a vacuum. It is conducted with full cognizance of the relevant investigations conducted by other scientists, using other methods, in such fields as marriage and the family, labor and management, intergroup, and international relations. The present paper does not permit even a brief summary of the broad empirical background within which the experimental work proceeds. The general strategy of the latter approach is to try to capture the essential properties of important types of interpersonal relationships and, through exploitation of the special opportunties for control and analysis which the experimental method provides, to identify in detail the various types of accommodative processes and the factors that determine their success or failure. Given this detailed analysis, observers of natural and more complex matters will be able more easily to dissect and understand their phenomena and to formulate policy recommendations regarding means of minimizing fruitless conflict and increasing the quality of achieved accommod-tions.

ACKNOWLEDGMENT

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IV. An Approach to Bioengineering

Y. C. B. FUNG

INTRODUCTION AND HISTORICAL BACKGROUND

WHAT IS BIOENGINEERING? Obviously, bioengineering is the inter-
section of biology and engineering. Biology is the science of life.
But what is engineering? It is often debated among engineers. Re-
cently, several societies held long discussions on the goals of engi-
neering, and a definition that evolved is that "Engineering is the
search for and use of scientific knowledge to the benefit of man." It
may not be the definition accepted by everybody, but it describes
well the activities of most engineers. In this concept engineering
differs from pure science by the motivation toward application. The
difference between a scientist and an engineer is largely psychologi-
cal, and many people play the role of both. Thus, for engineering,
the method is scientific, the mode is quantitative, the dictum is
economy, the concern is human.

Bioengineering represents engineering directed at the living world.
It encompasses the search for and use of scientific knowledge in
biology for the benefit of man. It is also biology directed toward
engineering. A bioengineer can therefore be an engineer concerned
with living matters or a biologist motivated by applications.

The field of bioengineering is immense. Obviously, it includes
the engineering application of biology, and the biological applica-
tion of engineering. Some of the popular items are listed below. In
Table I are the engineering applications of biology. Bionics, the
imitation of nature in electronic systems, is destined to be an im-

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in 1966, Dr. Fung was a member of the faculty at the California Institute
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TABLE I

Engr. Applications of Biology

```
                    Engr. Applications of Biology
                    |
        ┌───────────────────────┬───────────────────┐
    ┌────────┐                            ┌──────────┐
    │ Bionics│                            │  Appl.   │
    └────────┘                            │ Biology  │
        │                                 └──────────┘
        ├─ Electrical                           │
        │  systems              ┌───────────────┼───────────────┐
        ├─ Mechanical       ┌──────────┐  ┌──────────┐  ┌──────────┐
        │  systems          │ Industrial│  │  Appl.   │  │  Agri.   │
        ├─ Miniaturization  │   Engr.   │  │  Psych.  │  │  Engr.   │
        │                   └──────────┘  └──────────┘  └──────────┘
        ├─ Memory                  │
        │                          ├─ Fermentation
        ├─ Sensing                 │
        │                          ├─ Filtration
        ├─ Locomotion              │
        │                          └─ Desalination
        └─ Computing
           systems
```
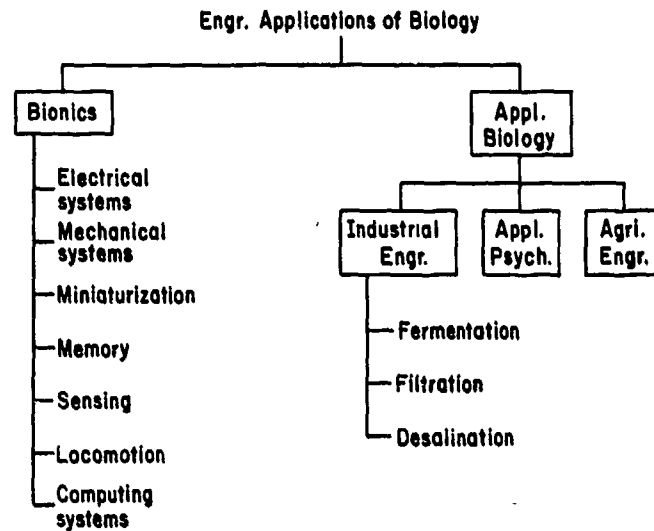
portant industry of the future. Fermentation, wine making, leather making, etc., are as old as our civilization. Filtration, purification of water with biological material, etc., are other examples. In Table II are some of the biological applications of engineering. We can think of an unending list of subjects useful to medicine, astronautics, and environmental health. Any one of these subjects is a big field. But on top of all these is a unifying theme: the enlargement of engineering principles to deal with biomedical problems. See Table III. This is the basic research which accumulates the broad, yet detailed, quantitative knowledge and understanding which makes any applications possible, safe, confident, economical, and elegant.

The rest of this paper will be devoted to the illustration of the main theme: the enlargement of engineering principles for biomedical applications. I shall take my examples from the problem of blood circulation.

Figure 1 is a schematic diagram of the human circulation system. The heart is the prime mover. The arteries and veins are the conduits. The capillaries are the irrigated fields where cells of the body obtain their food. Every cell in the body must live near a blood capillary. On the average a cell cannot live if it is farther than

about one-thousandth of an inch away from a blood capillary. Thus Krogh (1922) gave the classical estimate that the logical length of the capillaries in the human body is about 60,000 miles, and the total

TABLE II

Biomedical Applications of Engineering

Medical Engr.

- Biomechanics
- Prosthesis
- Artificial organs
- External handling of blood, tissue, and organs
- Radiation therapy
- Rehabilitation
- Medical Instrumentation
- Hospital planning
- Systems Analysis for assistance to diagnosis, surgery, treatment and research
- Aerospace medicine
- Bioelectronics
- Pharmacological testing
- Hypo and hyperbaric effects on living systems

Environ. Health

Closed Systems
- Bio-astronautics
- Space exploration
- Ecological systems

Open Systems
- Sanitary engineering
- Air pollution
- Water pollution

TABLE III

BIOENGINEERING

Basic Studies: Enlargement of Engineering Sciences to Deal with Biomedical Problems

Engr. Applications of Biology
- Bionics
- Appl. Biology

Biomedical Applications of Engineering
- Medical Engr.
- Environ. Health
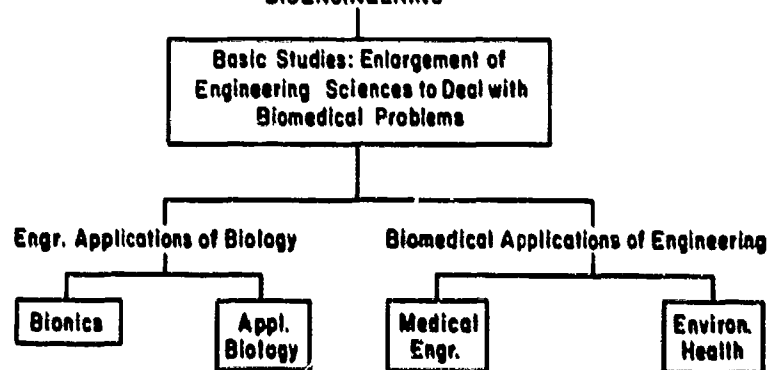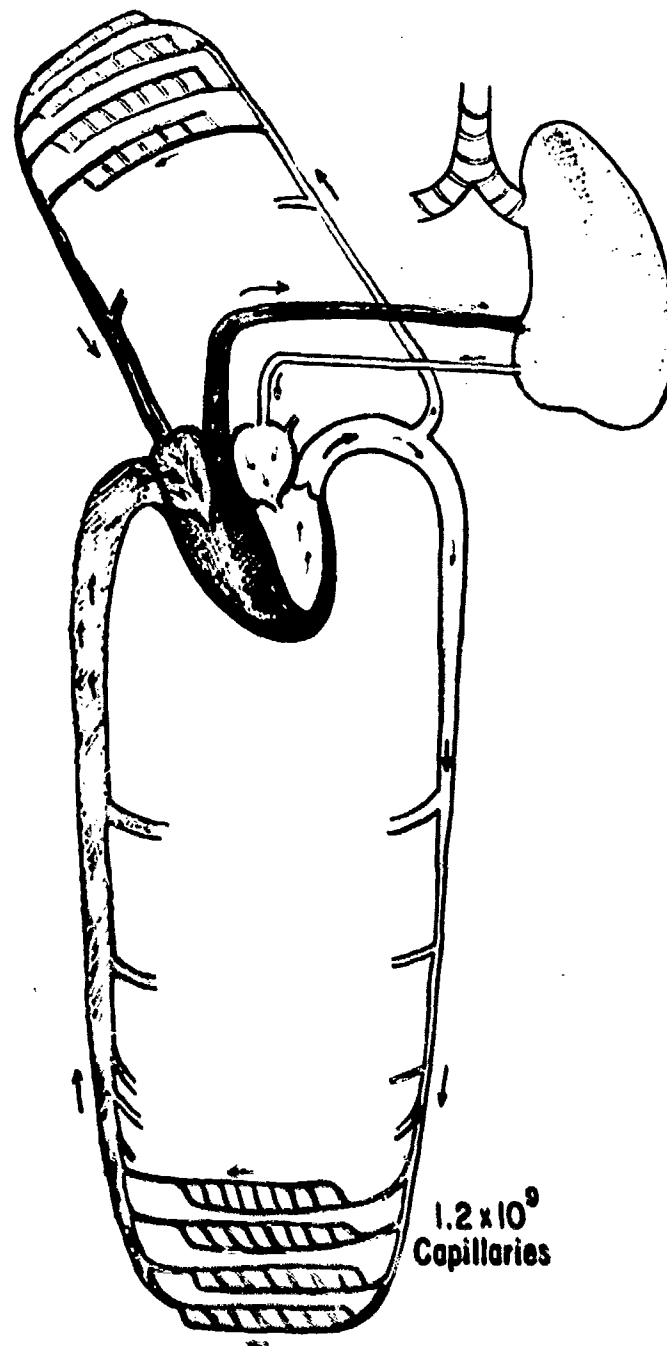
FIGURE 1. Schematic diagram of human circulation system.

surface area is 6,300 m². Our well-being certainly depends on the function of the capillaries. The brain, the heart, the muscle, all depend on the capillary blood vessels to live.

In a movie of blood flowing in the mesentery one can see the arteries, arterioles, sphincters, capillaries, venules, veins and the red blood cells. The red cells whiz by in the capillaries at a speed of about 1 to 2 mm per second. These red blood cells are very flexible. They assume all kinds of shapes in their course through the blood vessels.

Now, there are many problems associated with these flows whose solutions require advanced continuum mechanics. A number of the greatest masters contributed to this field. For example, in Table IV I compiled a list of names, which looks like one lifted from the history of engineering mechanics.

William Harvey, of course, is credited with the discovery of blood circulation. He achieved this discovery in 1616 by many critical observations and by logical reasoning. Having no microscope, he never saw the capillary blood vessels. This should make us appreciate his reasoning power even more deeply today because, without the capability of seeing the passage from the arteries to the veins, the discovery of circulation must be regarded as "theoretical". The actual discovery of capillaries was made by Marcello Malpigi (1628-1694) in 1661, forty-five years after Harvey made the capillaries a logical necessity.

A contemporary of William Harvey was Galileo. You remember that Galileo was a student of medicine before he became famous as a physicist. He used his pulse to determine the constancy of the period of a pendulum, and then used the pendulum to measure the pulse rate of people, expressing the results quantitatively in terms of the length of a pendulum synchronous with the beat. He invented the thermoscope and was also the first one to design a microscope in the modern sense in 1609, although rudimental microscopes were first made by J. Jansen and his son Zacharias in 1590. Robert Boyle studied the lung, and discussed the function of air in water with respect to fish respiration. Robert Hooke gave us the Hooke's Law in mechanics, and the word "cell" to biology to designate the elementary entities of life. His famous biological book, "Micrographia" (1664), was reprinted recently by Dover Publications. Leonhard Euler wrote a definitive paper in 1775 on the propagation of waves

in arteries. Thomas Young, who gave us the Young's Modulus, was a physician in London. He worked on the wave theory of light while he was concerned with astigmatism in lenses and in color vision. Poiseuille, while he was a student, invented the mercury manometer to measure the blood pressure in the aorta of a dog and discovered Poiseuille's law of viscous flow upon graduation.

Table IV

A List of Early Contributors to Biomechanics

Galileo Galilei (1564-1642)

William Harvey (1578-1658)

Robert Boyle (1627-1691)

Robert Hooke (1635-1703)

Leonhard Euler (1707-1783)

Thomas Young (1773-1829)

Jean Poiseuille (1799-1869)

Herrmann von Helmholtz (1821-1894)

Adolf Fick (1829-1901)

Diederik Johannes Korteweg (1848-1941)

Horace Lamb (1849-1934)

Otto Frank (1865-1944)

Balthasar van der Pol (1889-   )

To von Helmholtz might go the title "Father of Bioengineering". He was a professor of physiology and pathology at Königsberg, professor of anatomy and physiology at Bonn, professor of physiology at Heidelberg, and finally professor of physics in Berlin (1871). He wrote his paper "Law of Conservation of Energy" in barracks while he was in military service, fresh out of medical school. His contributions ranged over optics, acoustics, thermodynamics, electrodynamics, physiology and medicine. He discovered the focusing mechanism of the eye, and following Young, formulated the three-color theory of color vision. He invented the phakoscope to study the changes in the lens, the ophthalmoscope to view the retina, the ophthalmometer for measurement of eye dimensions, and the stereoscope with interpupillary distance adjustments for stereo-vision. He studied the mechanism of hearing and invented the Helmholtz resonator. His theory of the permanence of vorticity lies at the very foundation of modern fluid mechanics. His book "Sensations of Tone" is popular even today. He was the first to determine the velocity of the nerve pulse, giving the rate 30 meters per sec., and to show that the heat released by muscular contraction is an important source of animal heat.

The other names on the list are equally familiar to engineers. The physiologist Fick was the author of Fick's law in mass transfer. The hydrodynamicists Korteweg (1878) and Lamb (1898) wrote beautiful papers on wave propagation in blood vessels. Frank worked out a hydrodynamic theory of the heart. Van der Pol (1929) wrote about the modeling of the heart with nonlinear oscillators, and was able to simulate the heart with four Van der Pol oscillators to produce a realistically-looking electrocardiograph. The list would become too long if we were to continue further; it is perhaps sufficient to show that there were, and of course are, people who would be equally happy to work on the living as well as the nonliving subjects.

THE RED BLOOD CELL—AN EXAMPLE OF THE APPLICATION OF ENGINEERING MECHANICS TO BIOLOGY

I shall now outline a problem which we worked on recently. This is concerned with the most important unit of the circulation—the

red blood cells. These cells contain hemoglobin, which perform the task of oxygenation—taking up oxygen in the lung and passing it to the tissues of the body. When the red cells float in a stationary bath under the microscope, they appear as biconcave disks. At the top of Figure 2 are shown two views of a red cell of a rabbit under a microscope. The diffraction pattern was caused by the finite thickness of the red cell and by interference of light whose wave length (0.55 micra) was about 1/7 the radius of the red cell. The typical shape—a donut without a hole, is unmistakable. Viewed as structures, they are thin-walled shells with a radius-to-thickness ratio of order 400. The red cells of many mammals are about the same size. Human red cells have no nuclei, hence they cannot divide and multiply. The dimensions of a human red cell in unit of micron (10^{-4} cm) are shown in Figure 3. The red cells are very flexible. The lower photograph in Figure 2 shows the deformation of red cells in flowing through a capillary in the mesentery of a rabbit.

The question is, Why are the red cells biconcave? What consequences does the biconcavity imply?

We can never be too certain why red cells look that way They get that way in the process of evolution. The red cells of fishes are not biconcave, they have nuclei, and they are larger. Animals shed the nuclei of their red cells when they reach the stage of mammals. In adult humans, our infant red cells in the bone marrow have nuclei, but they expel their nuclei when they reach maturity, and only those biconcave red cells without nuclei are allowed to enter the blood circulation. It is easy to believe that something good must come from that particular geometry.

To see what consequences the biconcavity may imply, we must know the state of the contents of the red cells. Is it in the state of a viscous liquid, or is it a gel solid? By definition, we call a material solid if it can sustain shear stress statically without flow. To determine whether a material is a solid or not we must put some load on it and observe its deformation. The existence of an X-ray defraction pattern alone cannot determine the physical state because we know there are liquid crystals and there are plastic crystals. We know a great deal about the chemistry of the hemoglobin, but we do not know its physical state in the red cell. This simple question of deformability cannot be answered chemically, nor by an electron microscope, which requires fixing (and killing) the cells.
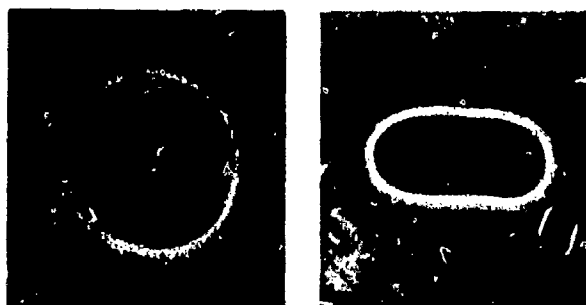
FIGURE 2. (Top). Photograph of a red blood cell. (Bottom). The deforma-
tion of red cells in flowing through a capillary blood vessel in the mesen-
tery of a rabbit.

AXIS OF SYMMETRY

1.3

1.0±0.08

2.4±0.13

8.5±0.41

AREA = $163\mu^2$, VOLUME = $87\mu^3$
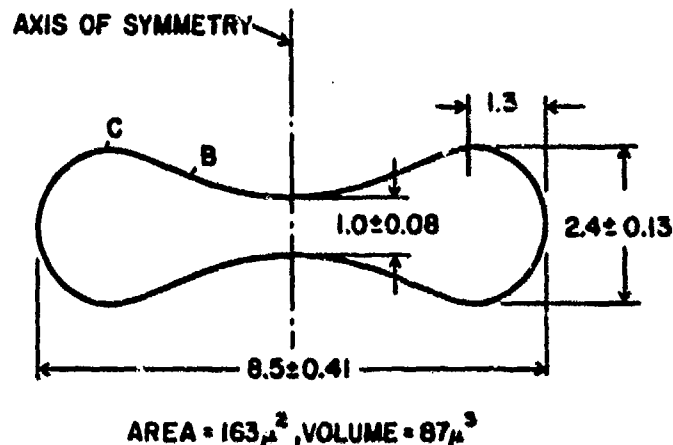
Figure 3. Dimensions of human red blood cell according to E. Ponder.

An indirect approach must be taken. Let us make two alternative assumptions. First, assume that the interior of the red cell is a liquid and deduce the mechanical properties of the red cell as a whole. Then we assume that the interior is a gel and see what differences in mechanical properties may be found. Finally, we compare the theoretical results with all the known experimental results and, if necessary, perform new experiments to make a choice. The knowledge gained in this process will be useful for further applications.

Assuming a liquid interior, we see a red cell as a thin-walled shell filled with a liquid. To analyze such a shell is similar to the analysis of a liquid-fueled rocket; except the red cell problem is more difficult mathematically. A thin-walled shell resists external load by "membrane stresses", i.e., the stress resultants in the shell wall. Now, if we ignore the variation of stresses across the thickness of the very thin wall, we find that the partial differential equation which describes the stress distribution in the shell is either of elliptic type or of hyperbolic type, depending on the sign of the Gaussian curvature of the shell surface. The differential equation is elliptic if the Gaussian curvature is positive; it is hyperbolic if the Gaussian curvature is negative. The Gaussian curvature is the product of the two principal curvatures of the surface. For a biconcave shell as illustrated in Figure 3, the Gaussian curvature is positive in the neighborhoods of both the equator and the poles but is negative

between the point of inflection B and the crown C. Therefore the differential equation for the membrane stress changes from elliptic type near the axis of symmetry, to hyperbolic type beyond the point of inflection, and back to elliptic type beyond the crown C. At B and C the Gaussian curvature is zero. Those familiar with the aerodynamic theory of supersonic flight would recognize that our mathematical feature is exactly the same as that of the sonic line in a mixed supersonic and subsonic flow. We have all the difficulties of the transonic flow theory.

Nature takes a dim view of mathematical difficulties. In transonic flow, shock waves soon appear. In our red cell theory, the features that come to the rescue are the bending and the nonlinear effects, leading to the buckling of the thin shell.

If there is a pressure differential across the red cell membrane, and if there is no buckling, then we shall encounter a singularity of infinite membrane stress at the crown. This can be demonstrated by considering the equilibrium of a segment of the red cell membrane as shown in Figure 4. Here A is the axis of symmetry. The
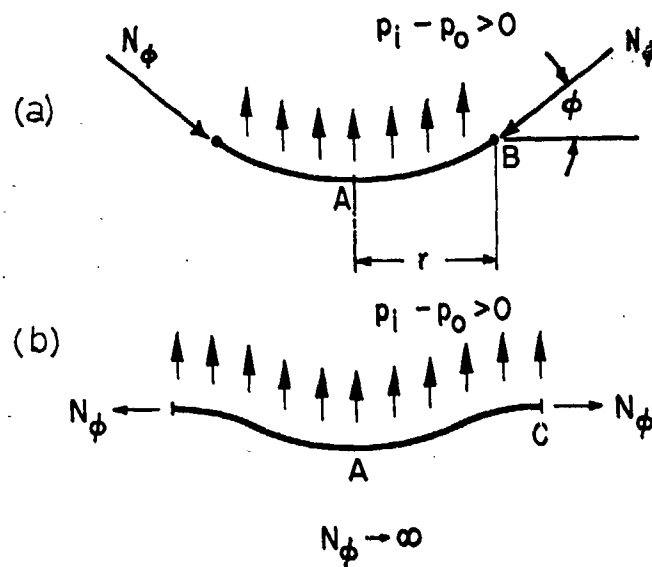
$$\pi r^2 (p_i - p_o) = 2 \pi r \, N_\phi \sin \phi$$

FIGURE 4. Equilibrium of a polar segment of the cell membrane.

pressure inside the red cell is denoted by P_1, while the outside pressure is denoted by P_o. If the pressure differential $P_1 - P_o$ is positive, the situation is as shown in Figure 4. The vertical resultant force $(p_1 - p_o)\,\pi r^2$ must be balanced by the membrane compression $2\pi r N_\varphi \sin\varphi$, where φ is the angle between the tangent to the membrane and the equatorial plane. Now at C the membrane is parallel to the equatorial plane, so that $\varphi = 0$, and N_φ must tend to infinity if $p_1 - p_o \neq 0$.

Perhaps no biological membrane can develop such an infinitely large stress. What actually happens when the pressure differential $p_1 - p_o \neq 0$ is that the bending and the nonlinear effects take over, the shell buckles, and the normal biconcave shape will be lost.

We are all familiar with buckling of thin shells under negative pressure, but it is equally important to remember that a thin shell of the biconcave type can buckle also under a positive pressure. It is easy to show by model experiments that the equatorial region of a biconcave shell buckles into a number of lobes as the internal pressure is increased and the pole region bulges out. This buckling occurs because when the cell tries to become spherical as the internal pressure increases, the equator must be sucked in. The buckles merely compensate for the reduction in equatorial length. This very simple reasoning, however, shows that the stress pattern in the red cell membrane must be quite complicated under such a condition.

We have concluded that if the interior of the red cell is in a fluid state, and if the pressure differential across the cell membrane does not vanish, then either the membrane stress N_φ would tend to infinity at the crown, or the cell would buckle. But the normal red cells in the normal living condition are seen under the microscope to be perfectly happy in the normal biconcave shape, without tearing and without buckling. Therefore, we have to reject either or both of the hypotheses; namely, the liquid interior and the finite pressure differential assumptions. Sidestepping the solid interior hypothesis for the moment, we conclude that if the red cell interior is a viscous fluid, then the pressure differential cannot be finite. Thus

$$p_1 - p_o \doteq 0 \tag{1}$$

or, more accurately,

$$p^{(-)}_{cr} < p_1 - p_o < p^{(+)}_{cr} \tag{2}$$

where $p_{cr}^{(+)}$, $p_{cr}^{(-)}$ are the critical buckling pressures under positive and negative internal pressures respectively. The calculation of these buckling pressures is not easy, but an estimate for the mammalian red cell is that $p_{cr}^{(+)}$ is of the order of 1 mm of water, $p_{cr}^{(-)}$ is of the order —0.2 mm water. Thus the internal pressure in the red cell in the normal, biconcave configuration is essentially zero.

When we look closer at the mechanics of the red cell, we find that there are other important assets associated with the biconcave geometry. One is the great deformability; the other is the reduction of stresses in resisting the wear and tear experienced by the cell in its course through the body, squeezing through the capillaries, turning the corners, getting caught in the sphincters, etc.

To explain this achievement let us look again at the whole cell. Let us ignore the bending stresses, which are small even at large changes of curvature because of the very small wall thickness, which is estimated to be 70 to 100 Angstrom. A deformation will induce no membrane stress if all the elements of length on the mid-surface remain invariant during the deformation. In differential geometry such a deformation is said to be *isometric*. A surface is said to be *applicable* to another surface if one can be deformed into the other by continuous bending without tearing and without stretching. Deformation of a surface into an applicable one is isometric, inducing no change in the membrane strain or stress.

It is well known from differential geometry that if two surfaces are applicable to each other, their Gaussian curvature must be the same at the corresponding points. Applying this rule we can determine whether a given shell can be deformed isometrically into another or not. For example, a developable surface is applicable to another developable surface. Thus a thin-walled cylinder can be deformed into a diamond-patterned bellows composed of flat triangles, as the reader can easily verify by rolling a sheet of paper into a cylinder and crushing it longitudinally by loading its ends. A sphere is applicable to another spherical surface by an inversion, as shown in Figure 5. But in either case the total volume enclosed in the shell changes during buckling. If the enclosed volume is not allowed to change, as in the case of the red cell which is filled with an incompressible fluid, these deformations cannot occur without accompanying stretching of the membrane. Here we realize the beauty
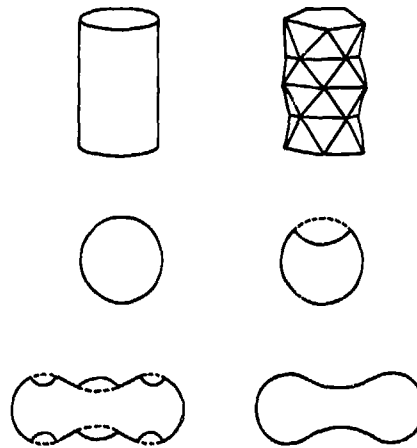
FIGURE 5. Example of applicable surfaces obtainable by isometric transformation. The red cell can deform isometrically and isochorically, so that both the volume and the surface elements are unchanged.

of the biconcave geometry of the red cell. There exist an infinite number of surfaces applicable to the normal geometry (see Figure 5). The red cell can deform into any one of these without any tearing and stretching of its membrane.

This at once explains the great deformability of the red cell and the soundness of the cell design. The role it plays demands that it be tough and flexible. It obtains the toughness by being able to deform easily and avoiding the stress. Of course, the red cells are stressed whenever the velocity gradient and the geometric conditions do not permit them to deform isometrically. These "off-design" conditions must be investigated separately in each special case.

We may continue to examine what happens when the volume of the red cell does change as when the cell is placed in a hypotonic or hypertonic medium; or what happens when the surface tension or the membrane structure is changed when certain chemicals are added to the plasma. We may look into what is to be expected of a ruptured red cell, the so-called *ghosts*. Finally, we may examine the consequences of the alternative hypothesis—namely that the interior content of the red cell is a gel solid. Analytic solutions to these problems are important because they offer us the means to determine

the red cell's properties: such as the distribution of the thickness of the cell membrane, its stress-strain law, its strength, its changes in flow in vivo and in vitro, and so on.

The theoretical results must be compared with experiments to see whether the hypotheses are justified. Our theoretical examination has led to a number of experiments which are still under way. At this time, the assumption that the normal red cell interior is a liquid seems to be in agreement with all known facts.

One may ask, "What is the use of such an investigation?" Our answer is, "We want to know how to handle the blood in the body and in the heart-lung machine. We want to know the causes of damage to the red cells and how to avoid them. We want to know how oxygenation is accomplished and how to control it. For these we want to know how the red cell is stressed, how it is damaged, how it interacts with the capillary wall, how it influences the blood viscosity, and so on." Our interest in the red cell mechanics is more than academic.

THE STRESS-STRAIN-HISTORY LAW OF SOFT TISSUES— INTRODUCTION OF A NEW MECHANICS

The red cell problem considered above illustrates the type of approach to biology that an engineer takes. To illustrate the other side of the interaction—the enrichment of engineering science by biology, it is perhaps best to consider the properties of materials that are thrust upon the engineer. Again consider mechanics. In engineering mechanics we are familiar with linear elastic materials which obey Hooke's law—that the stress tensor is a linear function of the strain tensor. This law describes admirably well most structural materials below the yielding point. Beyond the yielding point most metals show the phenomenon of plastic yielding. Under repeated-stress cycles most structural materials dissipate energy and deviate from Hooke's law, leading to the so-called viscoelasticity phenomenon. In recent years rubbery materials became important and the theory of finite deformation became a strong new branch of mechanics. In bioengineering, we have to consider living tissues. There are body tissues that may be described by these familiar laws. However, there are other tissues that require an entirely new description.

Let us consider the soft tissues of our body, such as the skin, the muscle, the blood vessels, and other connective tissues. It is obvious that the mechanical properties of these tissues are important to problems of physiology, pathology, medicine, diagnosis, and prosthetics. We shall discuss in particular the properties of the mesentery, which is a thin membrane connecting the intestines. The mesentery is a favorite tissue of the physiologist; it is transparent, of uniform thickness, and has a microcirculation system that can be easily seen under the microscope. Much of our knowledge about microcirculation is obtained from the mesenteric flow.

It is well known that smaller blood vessels are stiffer. The principal reason for this stiffness variation is simply the effect of the size. For geometrically similar blood vessels, the stiffness, defined by the derivative dp/dR, where p is the internal pressure and R the lumen radius, increases as R decreases. However, for the capillary blood vessels in the mesentery, the experimental results of Baez and Lamport indicate that the capillaries with diameter of order 10 micra are much more rigid than the geometric factor alone would imply. To explain this unusually high rigidity of the capillary blood vessels in the mesentery, we ask whether the tissue surrounding the blood vessels contribute significantly to the elasticity of the blood vessel. In the mesentery the capillaries appear to be embedded in a gel. However, it is obviously difficult to measure the distensibility of blood vessels both with and without the surrounding tissues. Hence it is not surprising that few data can be found concerning this subject.

One way to evaluate the effect of the surrounding tissue is to measure the elasticity of the tissue and then calculate the contributions under the hypothesis that the blood vessels are in direct contact with the tissue. For this purpose a series of experiments was made on the avascular portion of the mesentery of the rabbit. When these experimental results were applied to the mesentery, it was shown that the percentage contribution of the surrounding tissue to the total stiffness of the blood vessels is, for capillary, 99.7%, centrally located venule, 61.3%, arteriole, 45.2%, eccentrically located venule, 41.7% and terminal artery, 11.8%, provided that certain typical dimensions and typical stretching of the mesentery were assumed. Although these percentage values would vary if the assumed dimensions and stretching were varied, it is evident that

the surrounding tissue contributes greatly to the elasticity of the blood vessels. The larger the effective size of the surrounding tissue relative to the vessel, the greater is the contribution. The capillary, which derives almost all of its elastic stiffness from the surrounding tissues, may be described mechanically as a tunnel in a gel.

The analysis that leads to the tunnel-in-gel concept of capillary is an application of the stress-strain law of the mesentery. Let us now turn to the law itself. If a narrow rectangular strip of the avascular portion of the mesentery of a rabbit was tied at the ends and tested in a tensile testing machine, the results may be presented in Figures 6 to 8.

Figure 6 shows the load-deflection curve of a specimen when the rate of strain imposed was 0.254 cm per minute. The ordinate shows the load in grams. The abscissa shows the deflection in centimeters. The relaxed length of the specimen was $l_0 = 1.22$ cm. When the specimen was stretched from l_0 to $l_1 \doteq 2.54$ cm, the corresponding
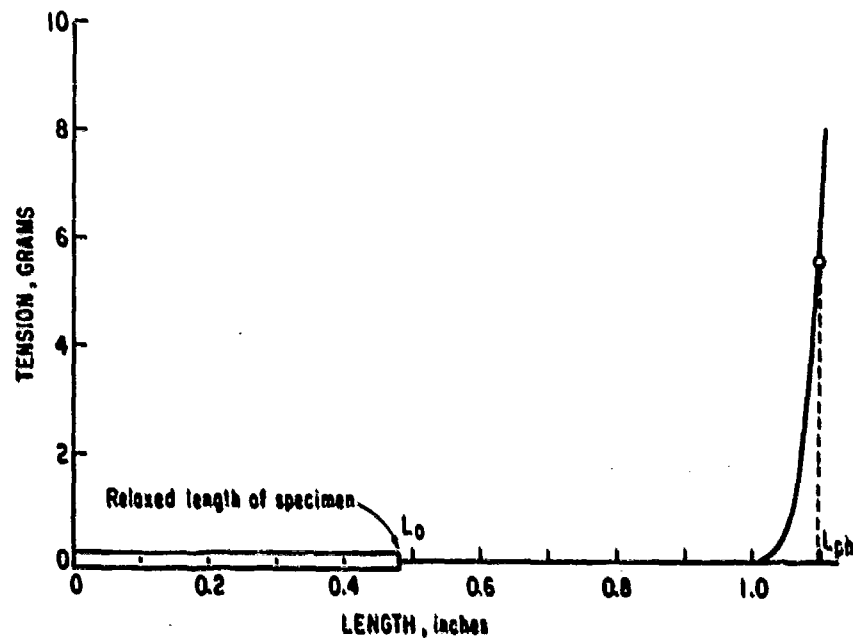
FIGURE 6. The load-deflection curve of a rabbit mesentery in tension. The state corresponding to the naturally spread-out mesentery is marked by the small circle. The point l_0 marks the relaxed length of the specimen.

tension induced was very small; in fact it was not readable in the chart illustrated in Figure 6. Extension beyond l_1 however, induced a rapidly increasing tension. The load-deflection relationship was definitely nonlinear.

Figure 7 shows typical hysteresis curves of the specimen. It is seen that hysteresis exists, but it is not very large. Although it is not shown
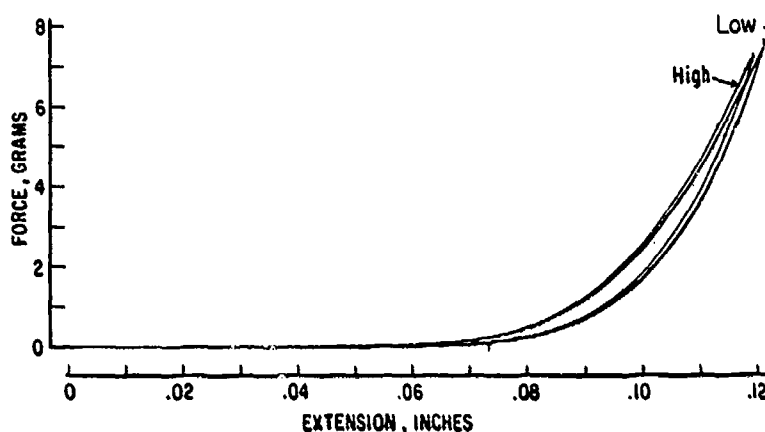
III - 6 RABBIT MES.
$L_0 = 0.34"$ $L_{ph} = 1.09"$
STRAIN RATE:
  UPPER, HIGH: ± 0.2 in./min.
  LOWER, LOW: ± 0.02 in./min.

FIGURE 7. Hysteresis curves of rabbit mesentery obtained at different strain rates. The high rate was ten times that of the low rate. Only slight change in hysteresis curves was obtained. Some of the small difference is due to fatigue, some is due to strain rate.

in the figure, a completely unloaded specimen gradually returned to the initial length l_0. In other words, there was no doubt that the material was elastic, although the modulus of elasticity was very small in the range l_0 to l_1. The curve marked "high" was produced at a strain rate ten times faster than that marked "low". It is seen that the hysteresis loops did not depend very much on the rate of strain.

Figure 8 shows a stress-relaxation curve. The specimen was strained at a constant rate until a tension T_1 was obtained. The length of the specimen was then held fixed and the change of tension with time was plotted.
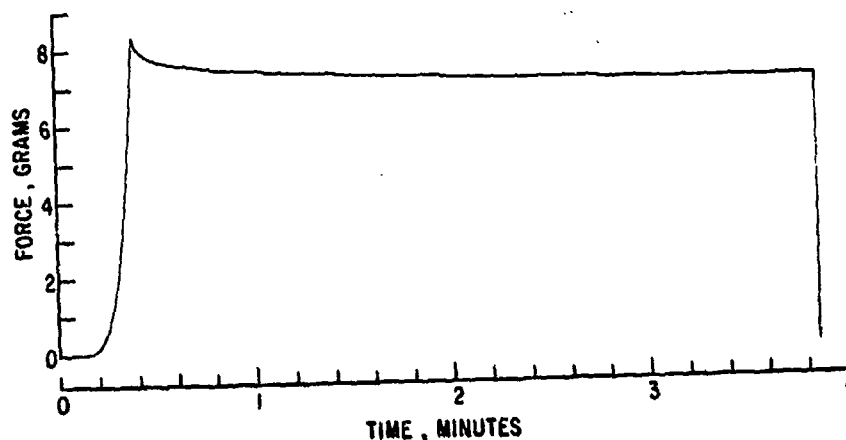
III – 3 RABBIT MES.
L_o 0.34"
L_{ph} 1.09" W_{ph} 0.394"
RATE TO PEAK 0.5 in/min

FIGURE 8. Relaxation curve. Rabbit mesentery. The specimen was stressed at a strain rate of 1.27 cm per minute to the peak. Then the moving head of the testing machine was suddenly stopped so that the strain remained constant. The subsequent relaxation of stress is shown.

There are other features of interest. For example, if the specimen is loaded and unloaded at a fixed rate of strain between two fixed limits of extension, the peak response decreases with the number of cycles. It is a form of "fatigue". Finally, if the specimen is strained up to failure, the failure curve resembles that of ordinary structural materials. The ultimate load for the specimen shown in Figure 6 was about 31 grams. The ultimate strain at breaking point was $\lambda = 2.17$. The specimen failed by tearing at some unpredictable points. In all these experiments the specimens were suspended in a physiological solution at room temperature.

It is evident from the curves shown in Figures 6-8 that the stress-strain relationship for the mesentery and the arteries is nonlinear, that the stress does not depend on the strain alone, but also on the strain history. Let the stress-strain relationship be separated into two parts: an *elastic* part and a *history-dependent* part. The elastic part defines a unique stress-strain relationship, i.e., the "elasticity" of the material. The history-dependent part is time dependent; it

is related to the hysteresis, stress relaxation, creep, and other non-conservative phenomena. Thus we may write, for the simple elongation,

$$T(t) = F[\lambda(t)] + F'[\lambda(t\text{-}\tau); \ t, \ \tau] \tag{3}$$

where $T(t)$ is the tensile stress at time t referred to the undeformed state, i.e., the tension divided by the original cross sectional area, and $\lambda(t)$ is the extension ratio which describes the strain. The extension ratio is defined as the length between two points in the deformed state of the body divided by the distance between these two points in the undeformed body. $F[\lambda(t)]$ is a function of the extension ratio λ at time t; whereas $F'[\lambda(t\text{-}\tau); \ t,\tau]$ is a function of the entire history of the deformation, $\lambda(t\text{-}\tau)$.

Consider first the elastic response $F(\lambda)$. The most striking feature of the elasticity of a living tissue as seen from Figure 6 is the very small stress in response to large initial strain. In Figure 6 an extension up to about 100 percent of the relaxed length yields only a small, unmeasurable tension. However, for λ greater than 2 the stress rises rapidly, and indeed, exponentially. When the slope of the elastic tension-deflection curve, $dT/d\lambda$, is plotted against the elastic tension T, a remarkable correlation exists which may be fitted by a straight line in the first approximation:

$$\frac{dT}{d\lambda} = aT, \qquad (1 \leqq \lambda \leqq \lambda_y) \tag{4}$$

An integration with the boundary condition $T = T^*$ when $\lambda = \lambda^*$ gives

$$T = T^* e^{a(\lambda - \lambda^*)}, \qquad (1 \leqq \lambda \leqq \lambda_y) \tag{5}$$

This simple relation is remarkable, indeed. It shows that the tensile stress is an exponential function of the extension ratio. It may be shown that the skin, the series elastic element of the striated muscle and the heart muscle follow the same trend. It appears that the exponential type of material is natural in the biological world.

Modification of this formula to account for the curvature of the experimental curve of $dT/d\lambda$ vs T can be introduced easily. However, it should be noted that T, as given by the exponential function above, does not vanish unless $\lambda \to -\infty$. By definition, however,

we must have $T = 0$ when $\lambda = 1$, which defines the unstrained state. Hence a modification is necessary in order to account for this initial condition. This can be done in several ways. The simplest way is to introduce a polynominal factor that vanishes at $\lambda = 1$. Guidance for such a modification can be obtained from the general theory of elasticity. It can be shown that if a strain energy function $W(\lambda_1,\lambda_2,\lambda_3)$ exists, then for an isotropic incompressible elastic body subjected to simple elongation, the Lagrangian tensile stress is given by

$$T = 2\left(\lambda - \frac{1}{\lambda^3}\right)\left(\frac{\partial W}{\partial I_1} + \frac{1}{\lambda}\frac{\partial W}{\partial I_2}\right) \tag{6}$$

where I_1, I_2 are the strain invariants.

When $\lambda \to 1$, the zero factor must be of the form $\left(\lambda - \frac{1}{\lambda^2}\right)$ if the strain energy has no singularity at the undeformed state, i.e., if $\partial W/\partial I_1$, $\partial W/\partial I_2$ are finite and continuous at $\lambda = 1$. Adopting this zero factor, we obtain

$$T = \frac{T^*}{\lambda^* - \frac{1}{\lambda^{*2}}}\left(\lambda - \frac{1}{\lambda^2}\right) e^{\hat{a}(\lambda - \lambda^*)}, \qquad (1 \le \lambda \le \lambda_y) \tag{7}$$

Here the constant $\hat{a}$ no longer has the same simple meaning as a, the slope of the $dT/d\lambda$ versus T curve. However, the exponential factor in this equation is so powerful that as far as the mesentery is concerned, Eqs. (5) and (7) plot out to be almost the same curves, with only a slight difference between a and $\hat{a}$. For other soft tissues, the polynomial factor may play an important role and cannot be ignored.

We note that, in general, the resting configuration of a soft tissue in the body is not the unstrained state. People with the experience of cutting a major artery often find that the vessel shrinks away from the cut. The determination of the unstrained natural state is a difficult task in biological experiments.

According to these facts, it is elementary to point out that the usual practice in the physiological literature to present one number for the Young's modulus of elasticity for a living tissue is meaning-

less. The Young's modulus of a living tissue varies greatly with the stress, ranging all the way from almost zero to about 5×10^6 dynes/cm² for the mesentery. On the other hand the slope at the origin, a, and the curvature, $2ab$, of the $dT/d\lambda$ vs T curve are constant over the physiological range for many soft tissues. These parameters, together with a specific tension (or Lagrangian stress) T^* at a specific extention λ^*, completely characterize the elastic curve for the lower stress range. They are the candidates for data collection and data presentation.

Let us now consider the history-dependent part of the stress-strain relationship. In this respect, the most striking feature of the mesenteric response, as shown in Figure 7, is that the hysteresis curves are insensitive to the strain rate. Otherwise, the relaxation curve of Figure 8 would suggest that the relaxation function may be approximated by an exponential function. However, any finite sum of such exponents (discrete frequency spectrum) would imply a frequency dependence of the hysteresis loop. Therefore, the hysteresis and the relaxation curves taken together mean that a discrete relaxation spectrum is inadequate. It turns out that the inversion of the experimental relaxation curve to determine the frequency spectrum is mathematically a problem of inversion of Laplace transform on the real axis (the transform is real-valued and is known only for real arguments), and is a rather tenuous process. The same relaxation curve may be fitted approximately by several frequency functions, and a selection of the exact inverse would demand a numerical precision of the relaxation function unobtainable in experiments. However, the choice of the inverse can be based on the hysteresis curve. With such a reasoning, we formulate the stress-strain-history law as follows:

$$T(t) = F[\lambda(t)] + \int_0^t \int_0^\infty \alpha(\nu)\, e^{-\nu(t-\tau)}\, \frac{dF[\lambda(\tau)]}{d\tau}\, d\nu\, d\tau \qquad (8)$$

where $F[\lambda]$, the elastic response, is given by the right hand side of Eq. (7). The integral is a linear superposition of the past stress history; $\alpha(\nu)$ is the frequency spectrum, and is assumed to be a continuous function of the frequency ν. The function $\alpha(\nu)$ spreads out the characteristic frequencies so that the strain rate effect is spread out in such a way that the hysteresis becomes rate-insensitive in the practical range of strain rates. The "semi-linear" superposition,

which makes the stress depend linearly on the stress-history and nonlinearly on the strain-history, is a simplification without which the stress-strain law would be much more complicated.

If the elastic stress $F[\lambda(t)]$ is a step function, then $T(t)$ is the relaxation curve, and it is seen that $\alpha(\nu)$ is the inverse Laplace transform of $T(t)$.

Without further elaboration on the analytical form of the spectrum $\alpha(\nu)$ and the three-dimensional generalization of the one-dimensional stress-strain law, we can characterize a living soft tissue by saying that the elastic stress is essentially an exponential function of the extension ratio, and that the viscoelasticity is semi-linear, whose relaxation function has a continuous frequency spectrum. As far as I know, there is no engineering material that behaves in this way. Mechanics of such materials must be worked out anew. There is no catalog of existing solutions to boundary-value problems of such materials. Developments in this field would undoubtedly foster further advances in nonlinear mechanics.

These examples show that the theme of bioengineering as a scientific discipline—the enlargement of engineering sciences to deal with biomedical problems, is not empty. Engineers offer a new set of tools—the methods of engineering analysis and synthesis, to biology and medicine.

In return, the need to sharpen their tools to deal with new problems in bioengineering would certainly enrich engineering sciences. However, we are witnessing only the sprouting of the seeds; the flowering and fruition would await the efforts of the future.

V. The Organisation of
Living Memory Systems

J. Z. Young

It has been noticed that wise biologists are unwilling to answer
the question "What is life?" To the layman it might seem that this
should be the problem that most concerns biologists, but the lay-
man forgets that science has given up trying to answer such simple
questions as "what is heat," or "light," or even "matter." However
it is a sign of the growing maturity of biology that although we re-
fuse to say what life *is*, we can now give a rather precise and unam-
biguous definition of living organisms. They are very complicated
systems and the definition is long and rather dull. It begins by
saying that the systems are composed of some 16 of the 92 natural
elements, mainly of carbon, hydrogen, oxygen and nitrogen. It notes
that these are far from being the commonest elements in the earth's
composition but they are the four smallest elements in the Periodic
System that make stable electronic configurations by accepting 1, 2,
3 or 4 electrons (see Wald 1964). Moreover carbon, nitrogen and
oxygen are the only elements that regularly form double and triple
bonds. The four common bioelements are thus particularly "suited"
to form molecules and since the elements are small the molecules
are stable. Living organisms are in fact remarkably stable. We ex-
press this by saying that they conserve patterns of order, often for
many millions of years. To quote one outstanding example, the
Australian lung-fishes are almost identical with their ancestor
Ceratodus, which lived 300 million years ago. Again, the shrews that

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Biological Station at Naples. He became a Fellow of the Royal Society
in 1945.

are alive today are very little different from the shrews of the Cretaceous period 80 million years ago, who incidentally were also very like our own ancestors.

Besides containing much that is stable, living things are also very liable to change slightly, moreover they exist in great variety. This may be partly a reflection of the properties of the element carbon, with its four valencies and great families of homologous series of compounds, fatty acids, esters, alcohols, aldehydes and so on. Yet in spite of their variety all living organisms are built of remarkably similar molecules. There are compounds such as nicotinate, pantothenate, thiamin, adenosine triphosphate and many others that occur not only widely but probably in all animals and plants. Elaborate compounds, such as the porphyrins, turn up in slightly different forms as the chlorophyll of plants (combined with magnesium) or the haemoglobin or cytochrome of animals (combined with iron). Especially astonishing are the similarities of the proteins. These are composed of chains of amino acids, variously folded. But only about 20 of all the possible amino acids are used in animals, plants or bacteria. Most astonishing of all is the regulation of the order of living things by what we shall call the informational molecules, the nucleic acids. These serve to arrange the amino-acids in their particular orders on the protein chains. It is probable that approximately the same combinations of these nucleotide bases serve to introduce given amino-acids in viruses, bacteria and man. It is as if the instructions of the living systems were all written in the same language.

At more elaborate levels of organisation there are further similarities between organisms. Thus most organisms are based on the cellular structure of nucleus, cytoplasm and cell membrane. The electron microscope has shown a large range of organelles that are very widely spread among animals and plants. The endoplasmic reticulum is a system of intracellular channels, to some of which are attached the ribosomes which are concerned during protein synthesis to carry the amino-acids to their correct positions on the proteins. The mitochondria are folded structures carrying the enzymes concerned in making energy available. Cilia are little whips attached to cells to produce movement and they have almost identical structures wherever they occur, with a ring of nine fibres and two more in the centre.

With all of these facts, then, we can indeed define living organisms very much more precisely than was possible when all that could be said was that they were made of protoplasm or animal spirits. Nevertheless it will be felt that by such descriptions of the composition of organisms we have not come very near to achieving what could be regarded as an adequate definition of living things. And indeed it is characteristic of them that they are not entities at equilibrium and therefore cannot be statically defined. They are open systems, continually interchanging with the environment and maintaining a steady state. This active interchange is perhaps as near as we can come to an abstract entity that we can refer to as "life." The most characteristic thing about it is that it continues. The biologist thus must recognise the directional characteristic of the reactions that he studies. These reactions have had the effect of maintaining this astonishing continuity of living systems probably for over 2000 million years. The subject matter of biology demands an enquiry into how this stability is ensured. It is not only that the biologist is not ashamed to study this directional activity but rather that he is neglecting his subject if he does not do so. When a biochemist studies the reactions of the substances that he finds in an organism today, he will sooner or later discover that he has to study their history and indeed their function or purpose in the plan of self-maintaining activities.

This, of course, takes us back to the question of how such systems began. The origin of life is a question that is discussed by reputable scientists today and we can only say here that their tentative conclusion is that it began by the formation of complex compounds in a reducing atmosphere over 2000 million years ago. There is a widespread "belief" that this occurred by "natural" processes but there is doubt as to what these conditions might have been to account for the production of the particular ordered sequence of nucleotides that would serve to produce an active protein. There is, indeed, in this discussion an element of "which came first, the hen or the egg," since the nucleotides can only operate with the assistance of enzymes to build proteins, but the enzymes are themselves proteins. Haldane has speculated that the first self-replicating nucleic acid molecules might have been able to code for ribonuclease, which is one of the simplest of proteins (containing about 120 amino-acids).

Even the simplest self-maintaining organisms today, the bacteria,

are of a complexity that is very difficult for us to visualise or other-wise model. They maintain their active balance and steady state by putting into operation at the right rate some thousands of enzymes, which are catalysts that make reactions take place at relatively low temperatures. It is interesting to try to elucidate the principles by which all this activity is so regulated as to ensure continuity. We may concentrate upon two of them. First there must be a repertory of possible states of the system, incorporated in the various nucleotide combinations that have survived because they have made proteins that have been useful for the organisms in the past. The biochemist will find no explanation for the presence of any one nucleotide combination unless he knows that it codes for a particular protein that has played a part in keeping the system stable and may be needed again. This set of states thus constitutes the basic memory record. It is a representation of the past in the literal sense that it allows the organism to re-present to the environment a response that is appropriate for ensuring continuity.

The second principle thus emerges as the power to respond to the environment. This depends upon some form of detectors, sensitive, for example, to excess or to deficiency. Thus, in the classic case, a bacterium in the presence of a new raw material (substrate) will synthesise an enzyme to make that material available. Without go-ing into detail we can say that this depends upon having a) some detectors or sensors of environmental conditions, b) a communica-tion channel by which to transmit to the memory what we may call the information as to what is needed so that c) actions that are effective are performed. For bacteria Jacob and Monod and other biochemists have provided a lot of evidence about the details of this communication channel.

The fundamental principles for the maintenance of a stable steady state system are thus essentially those of communication. These include the presence of a selected memory record represent-ing events in the past, sensors able to detect change and to signal to the memory so that effective actions are performed. These are indeed obviously the minimal elements that must be involved in a steady state, but the logical features of the organisation of such a system are less clear than they seem. The actions must be such as are compatible with the future conditions of the surroundings and this is ensured only if the actions forecast the conditions. All life

participates in the character of what v. Foerster (1962) has called an inductive inference computer. Such activities are particularly conspicuous in the nervous system, with its memory and communication networks. But this is only a special case. We shall now show how the same principles of communication and control are manifest in all living activities and in this way we shall try to elicit the essential features of those principles.

The concept of "control" is not at first easy to reconcile with the physical scientist's approach to the organism as a reacting system like any other. It can perhaps be expressed by saying that the present state of any living system is dependent, like any other system, (1) on the surrounding conditions (2) on its present internal state, which in turn depends on its history. There is nothing unusual in this, except that the "informational" molecules are of a character such as to carry an unusually long and complicated record of their history. So we are back at the fact that the essential character of living organisms is their peculiar components, but we have added now that these lead them to operate in certain unusual ways, which give the appearance of foresight.

The particular state of a living organism at any time is thus dependent upon two sets of factors, the internal organisation that it receives from the past and the constraints imposed from outside at present. This *double dependence* ensures that the state and actions of the system are such as to lead to its continuance (Young 1946). Organisms have this property, of course, because selective action upon the memory in the internal organisation has insured that it "instructs" the system to proceed in that way, selecting "correct" responses from the repertory that has been built up. We have now to ask how living systems manage to make "correct" responses.

The capacity to make the right adjustments depends upon the dynamic characteristic of the system, on the fact that it is constantly changing. One of the most revolutionary of all techniques in biology has been the demonstration, by use of isotopes, of what Schoenheimer called the "dynamical state of bodily constituents." Tissues vary considerably in their degree of permanence, but in the majority there is a rather rapid turnover. Thus by injecting cholesterol labelled with ^{14}C into the yolk sack of day-old chicks it can be shown that this labelled material has a half-life in the liver or kidney of about 5½ days. Some constituents have shorter half-lives, others

longer; for instance, the collagen of the tendons once laid down turns over little if at all. Particularly important is the fact that there is a nuclear component that turns over very little. After labelling of rat's brains with tritium and later separation by centrifugation, it was found that about 20% of the material in the nuclei retained its activity for 6 months. This is about the proportion of the nuclei that is made up of DNA.

Great parts of the living system are thus continually changing and this gives them the opportunity to be adjusted to meet changing external circumstances. This continual replacement is the action system of the inductive inference computer. These continual homeostatic adjustments, as it were, compute the best state for the future in the light of the memory from the past and the present conditions. It is this that ensures against the risks of disruption by accident, which are inherent in any highly organised system. We have treated the organism as a planned device with mechanisms available to meet changes in its surroundings. These mechanisms include special repair processes ready to meet the types of damage to which the body is liable. We are so familiar with repair systems that we forget that they are as much part of the physiological repertory as, say, the actions of heart or lungs. Clotting of the blood is a particular case where the mechanism is known in some detail and we also have direct evidence from the hereditary disease, haemophilia, that it is genetically controlled. There is in fact a very large range of regenerative powers, for example in the skin, bones, nerves or liver. We are apt to think that mammals are limited in this respect compared with lower animals, which can regenerate whole limbs. But it is almost certainly a question of selection of those repair processes that are effective in keeping the individual alive or allowing it to reproduce. Mammals and birds do not regenerate limbs because if they lost one, in the period without it they would not be able to compete in the complex environments that they occupy, especially since they are warm-blooded and need constant supplies of food. Newts can float about in the water even after loss of a leg and still make good use of a new limb, when it grows. Frog tadpoles can grow new limbs, but not adult frogs, which would be too handicapped without them.

All these repair mechanisms are, in effect, special developments of the turnover processes, which in turn are special features of the

growth process to which we must now turn. The feature of living systems that produces all these adjustments is the tendency to replicate—to grow. It would be satisfactory if we could define this in terms of the capacities of the so-called self-replicating molecules of the nucleic acids. These have, indeed, remarkable properties of acting as primers and organisers of the synthesis of others like themselves. But they cannot, of course, do it simply on their own. They replicate only as parts of systems including other molecules, in particular those of the nucleotide polymerases. Though we cannot, therefore, say that we can yet fully define the chemical conditions of growth we can see them much more clearly now than was possible even few years ago.

Given suitable conditions, the mass of matter in a living yeast cell increases at a constant rate. Then after every four hours or so the rate suddenly doubles and the cell divides (Mitchison 1957). In a population of organisms such behaviour obviously leads to a logarithmic increase and this seems to be the basic rule for the synthesis of living matter. What has grown is capable of growing. This is the driving force lying behind all the adaptive characters of life. Of course, in organisms like ourselves the growth tendency is checked, so that instead of becoming mere unwieldly aggregates, the highly differentiated body is produced, with all its organs, each of the right size. But the growth power is there nevertheless and again becomes manifest in the repair processes such as those we have mentioned.

Indeed, the growth process never really stops; it only reaches the steady state of turnover, in which synthesis is matched by destruction. This is well seen in the many tissues, such as the blood, whose cells last only for a short time and are then destroyed. It is also conspicuous in tissues that are especially liable to damage, such as those of the skin or lining of the intestine, which are continuously replaced.

But, as we have seen, turnover continues in the majority of tissues, and turnover is a form of growth in which synthesis and destruction within the cells are balanced.

This continued renewal of the tissues is the mechanism that ensures their effectiveness as inductive inference computers, providing memory systems for producing continued survival. Like all planned systems those of the body are subject to random errors

and accidents of many sorts. Some accidents are bound ultimately to disrupt the functioning of any and every part and of the whole body. Repair processes are provided for the accidents to which each part is prone, but the repair processes themselves are liable to defect. Continuity is only ensured by repeated rejection and renewal of parts and ultimately of whole organisms.

It would be interesting to know more of how renewal is related to daily functioning and to the demands and stresses imposed by the surroundings. Many tissues have remarkable powers of adaptation, obvious examples being the growth of muscles with use and the increased oxygen carrying power of the blood induced by life at high altitudes. Presumably, here, there are receptor mechanisms which call upon specific synthetic response systems. But for us the interesting feature is that the muscle or the blood carry their memories of the past not as coded records or changes of instructions but simply as a change in their functioning capacity. They answer the question "Have you been as high up as this before?" simply by the way they act. We may find this a valuable clue when searching for the memory of the nervous system.

We begin to see then some sort of outline of the "plan", selected through the millenia, by which living things remain alive. It is a plan depending upon communication systems to produce adjustments at a series of time scales so as to ensure continuity. Higher organisms such as mammals have some very sensitive and rapidly adjusting systems, for example, those that regulate the heart beat, respiration and other processes that we ordinarily call physiological. The communication is here through the nervous system, which incidentally contains sections operating on different time scales, much slower for internal ("autonomic") than external ("somatic") events. Rather slower and longer lasting adjustments are made by the chemical signalling systems of the endocrine glands. The hormones are more suitable than nerve impulses to control adjustments that continue over a long period and involve changes at points with addresses in many parts of the body.

Still longer term adjustments are those adaptations we have been considering, by which muscles and bones grow strong with use or one kidney or other gland becomes larger if another fails and so on. These, like the simpler physiological adjustments, depend upon special mechanisms, allowed for in the instructions.

You will notice that as we proceed to longer time scales we pass to events whose occurrence is less likely. Changes in heart rate are certain to be demanded of every organism, as also are regulation of feeding and excretion. Hormonal mechanisms are called into play repeatedly, but irregularly, as in the stress responses of the adrenal gland, for example. Adaptive responses of muscle, blood, bones and glands are still less frequent and vary more with the activities of the organism and the accidents imposed by the environment. The repair mechanisms such as those for bone, are even less regularly called upon and the body, as we have seen, is only able to effect repairs that have reasonably often proved to be effective in the past.

All of these mechanisms together ensure survival, but only for a strictly limited period. If organisms depended only upon them, life would long ago have become extinct. The repair mechanisms gradually fail and senescence supervenes. In a state of nature, accident usually kills the organism before old age is manifest. Even apart from accident, repair mechanisms are likely to become less effective with age, if only because of the decreasing effectiveness of selection against characteristics that manifest themselves later (i.e. after some offspring have already been born). We cannot here enquire into all the complexities of the study of ageing, but one feature is of special interest, namely the production of errors in the instruction system of the DNA. These are bound to occur, perhaps especially as the molecules are used for copying. There is indeed some evidence that errors in the DNA are repaired. It is claimed that they are recognised by the "nonsense" molecules that they produce and are then removed and replaced with the information provided by the homologous strand (Falzone et al., 1966). It is said that the turnover of DNA increases in older people but there seems some doubt about this.

For our purposes the point is to emphasise once more the vulnerability to accident of any planned system. The continuity of life demands some better insurance than planned repair. And, of course, life has a better insurance. Continuity throughout the centuries is maintained by repeatedly submitting the instructions to the hazards of reproduction. Whenever a cell divides there is a period in which the genes are not operative. During that time only those metabolic systems that have previously been produced maintain the life of the cell. This acts as a filter for the instruction system that produced them. If the enzymes are ineffective because of a deficit in the in-

structions then that set will not be perpetuated (Falcone et al 1966). This selection is doubly vigorous during the haploid phase of sexual reproduction, when the insurance of the paired instructions is lost. It is interesting that in both plants and animals the main operating phases in which short-time adjustments are made use diploid and often polyploid instructions. These provide some protection against damage to the DNA. The haploid phase is a short and sharp period of selection. This may be the explanation for the vast production of sperms, since in these the organism is reduced to the instructions, together with the minimum of operating machinery. Abnormal sperms are certainly exceedingly common and the selection must be very severe, so that the metabolic machinery is tested to its limits and the DNA that produced it is passed on.

Reproduction, then, is the guarantee against error in the instructions. It is significant that no mass of living tissue continues indefinitely without division. But reproduction does more than this; it allows for the longest term adjustments of all, those of evolution. By mutation, selection and recombination it ensures that new sets of possible actions are available for selection according to the conditions of the environment. The long term changes that we call evolution are thus the continuation of the processes of adjustment that begin with the second by second adjustments of the heart rate. The mechanism of the adaptive change is similar from one end of the time scale to the other in that it depends on selection from sets of possible responses. On the evolutionary time scale the selection is not accomplished by feed-back through delicate sensors and communication channels, but by the harsh facts of survival. But such systems of random selection among a large number of slightly different possibilities allow some of the most precise adjustments (Platt 1958). Our astonishment at the accuracy and intricacy of living activities is a measure of our rating of their effectiveness as systems of planning.

Thus living systems make their forecasts on a great span of time scales. We can now return to examine in more detail the process of forecasting in the nervous system. Remembering that, although it is a particularly ingenious forecaster, it is only one among the many in the body and that the methods that it adopts are special developments of those that are used throughout. It has been emphasised that all parts of the body make their forecasts by continuously ad-

justing their action systems in the light of information received by some system of sensors of surrounding conditions. This feature of organisation as a communication channel is, of course, particularly characteristic of the nervous system. Indeed, when we speak of communication in the body we are apt to think of nervous communication rather than any other sort.

An especial feature of the nervous communication is that distant parts of the body are put into communication with each other, whereas the communications we have been speaking of hitherto have been from one part of a cell to another or among neighbouring cells. However, the blood and its hormones also provide a widespread system.

The signals carried in the nervous system are very precisely addressed. The network is laid down under the influence of heredity, controlling the operations of embryonic development. These provide nervous channels by which appropriate actions are taken to maintain life. First, as a child develops, by the simple processes of respiration, digestion etc., followed by more complex actions, sitting up, walking, talking and so on. The effectiveness of these is ensured by a great multiplicity of channels. In the nervous system each channel is the axon or output fibre of a nerve cell and it usually carries only information of one type. The signals are the nerve impulses, all alike in any one fibre, varying only in frequency and pulse distribution. These impulses propagate electrically in an all-or-nothing manner to the end of the nerve fibre. Here connection is made either with the receiving end of another nerve cell or with a muscle or gland. Single signals do not usually pass at these gates, the synapses. Mostly it requires a particular pattern of frequency in one fibre, or distribution of impulses in several incoming, pre-synaptic fibres to produce an outgoing signal in the post-synaptic cell. Moreover the transmission from pre- to post-synaptic is usually mediated, not electrically, but by secretion of a transmitter substance. These include amines, such as adrenaline, nor-adrenaline and 5-hydroxytryptamine, amino acids such as gamma aminobutyric acid and glutamine, and the choline ester acetylcholine. Some of these act as excitors, others as inhibitors of the post-synaptic cell. The balance of these effects constitutes, as it were, the act of computing by which the nervous system decides what actions are to be taken. Particular patterns of input in groups of afferent (sensory) nerve fibres produce

appropriate motor outputs. When you tread upon something sharp, the signals in the receptor fibres that are activated are so routed that shortening occurs in the flexor muscles of the leg that was pricked and in the extensor muscles of the other leg. Thus while one bends the other straightens—to hold the body up. Moreover, to produce the appropriate action of lifting the foot the relevant antagonistic muscles must be inhibited. The flexors cannot contract effectively unless the extensor muscles relax.

If the nervous system operated only with these principles the creature would move like a puppet, pulled by strings. In fact, the nervous system is not just a passive transmitter of signals. It contains many units that are continually active in more or less complex patterns. Some of these produce relatively simple rhythmic actions, such as those of breathing. Others, much more complex, maintain the extremely elaborate patterns of activity of the higher nervous centres. We understand relatively little of these activities at present, but it is essential to keep in mind that most of the cells of the brain are continually changing their thresholds, that is, their tendency to send signals. These changes occur either because such variations are built into the composition of the cells or under the influence of rhythmic excitation from central systems with specific activating functions.

So far, the actions of the nervous systems have been described as proceeding within an invariant network, developed under the influence of heredity. In reality no nervous system remains static in this way, even the simplest parts are modified by use. Probably it is by special developments of this power to be modified that there develops the memory system which is the object of our search.

Many facts indicate that nerve cells and fibres grow if they are used and atrophy and may die if they are left without input. Unfortunately, very little is known of the biochemical basis for these so-called trophic responses of nerve cells and fibres. This is one of the lacks that leaves us in a weak position for considering the more specialized memory processes that have presumably grown out of them.

The development of a memory facility depends upon the presence of alternative possible outputs following the stimulation of given input channels. Of course, even in a fully reflex system of response, activation of any given channel varies according to the other

channels with which it is stimulated. The response to treading on thumb tacks with both feet is not the same as with one. But for there to be an effective memory, there must be alternative possible action whose probability of future use can be altered in the light of the effects for the organism that have followed from their use in the past. This means that the memory system must be provided with some means for identifying what have been the results of the actions and this is, of course, one of the functions of signals such as those of taste, pain and other systems of reward. In our work with octopuses we have tried to work out the system by which these signals come to alter the probability of future actions. We suppose that there are classifying cells in the receptor system, each sensitive to some particular environmental change and that these cells may produce alternative actions. Thus when, for example, a horizontal rectangle appears in the visual field a horizontal classifier is activated and could make the octopus either attack it or retreat. In fact, an unfamiliar object excites a slow and cautious attack. Suppose the result is a shock, then the signals indicating that aversive behaviour is indicated must somehow not only activate the system for retreat but also increase the probability that retreat will follow when a similar rectangle appears again. This, it is suggested, may be the function of collateral channels leading from the retreat fibres and serving to block the "attack" pathway leading from that classifying cell, turning it into a cell ordering "retreat". The particular mechanism that we suppose may be operative is that these recurrent channels activate small cells with short processes that are specialized to begin to emit an inhibitory substance when they are appropriately activated. Such cells occur in the parts of the brain that are known to be necessary for the two separate memories of the octopus. One memory is for objects seen and another for objects touched. Moreover, in the touch system it has been shown that after removal of all these small cells the animals can no longer learn not to take an object that has been associated with pain (Wells & Wells 1957, Wells & Young 1966). There is, therefore, some evidence that these cells are associated with an inhibitory function. We even have some suggestion of how they do it. Electronmicroscopy shows that they are packed with the vesicles that have been shown almost certainly to contain transmitter substances. But these particular synaptic processes are peculiar in being in contact with other processes *also* containing synaptic

vesicles. The suggestion is that when the small cells are activated, say by signals of pain, they release a substance that blocks or inactivates the excitatory effects of the fibres with vesicles with which they are in contact. Such an action might be turned on suddenly by the activation of an enzyme system, and would then block the unwanted pathway indefinitely. We have, in fact, evidence that the octopus memory system shows a considerable persistence for up to four months, which is a large part of the animal's life. There is also evidence that, although the change once it has been made is not reversed, if the system is not tested by use it may seem partly to "forget", in the sense of making at first some wrong responses when tested with a given figure after a long time.

According to this view, then, the change that constitutes the memory consists in alteration of the probabilities of various actions as a result of experience of their results. More specifically the possible outcomes become reduced and limited. Situations that might produce one of several outcomes, later, as a result of experience, produce only one. This surely is not a bad description of the process of learning as we experience it ourselves. The particular feature of human memory is the capacity to react appropriately to a large number of particular detailed situations, especially of course, those concerned with speech. This brings us to the problem of whether the memory contains anything similar to an elaborately addressed record. We often have the experience that information is available if only we could find it. Yet it is quite unlikely that the items are filed in a highly classified system such as that of a computer. Each point in a computer record contains "information" only in the sense that when consulted at the correct moment as part of a program its "answer" will have some relevance to events in the outside world. Each point can be returned to a neutral state and used over again to represent another sort of information. Nerve cells are not like that. Each of them has a special relation to some particular type of event in the outside world. This is, indeed, the feature that gives to the nervous system its greatest asset—it can take data direct from the world because it has literally a certain homeomorphism with it. The receptor systems and brain constitute, as it were, a matrix for modelling the world. In its untaught state the matrix is capable of making various models, or to put it otherwise, the child can learn to live in any of many different worlds. Different, that is, in many

respects, though the choice is more limited than we suppose. To live a relatively effective life all people must walk, feed themselves with their hands, speak and behave in more or less conventional ways. We still do not know how much of our capacity to do these things depends upon the maturation of the nervous system under heredity and how much to later learning. The power to learn is certainly itself an inherited capacity. But there are considerable limitations on what we can learn, though our powers are much wider than those of any animal. We are not a perfect instrument, able to learn anything, but at birth our brains are more nearly blank sheets than those of any other animal. The essence of a good piece of writing paper is that it does not have anything written on it already.

A good computer can perform a wide variety of operations upon a range of numbers. Yet it may be that even our most complex powers, such as the use of language, depend in considerable detail on hereditarily determined patterns of brain action. The brain is an ingenious inductive inference computer, but one life is too short to allow the accumulation of the immense amount of information that is used in language. "The problem for the child is not the apparently insuperable inductive feat of arriving at a transformational generative grammar from restricted data, but rather that of discovering which of possible languages he is being exposed to" (Chomsky 1967). In other words having discovered that those around him are speaking Spanish the child at once preceeds to use the rules of Spanish grammar or rather the Spanish version of the universal grammatical rules.

Thus, as v. Foerster puts it, the analogy of a written record is not a happy one. The brain is not a great big reference library, in which items lie stored even if they are never referred to. It is a system for daily action, composed of a heirarchy of parts, though we know little of the physiology of how they operate. The actions it takes tend to perpetuate the individual and this tendency increases as time goes on and actions with unfavorable results are eliminated. This is a marvellous inductive inference computer, but unfortunately like all other machines it is subject to wear. It operates well for some three score years, with minor repairs, conducted as its substances turn over. By that time small defects for which there is no repair begin to accumulate and from there on we have to watch

a decline. The penalty for storing information in any one particular piece of matter is that it must deteriorate whether recorded upon a clay tablet or in a brain. Only by rejection of that information and return to basic operating instructions is the essential nature of the system preserved. But man has found a way around this difficulty too. By speech, and particularly by writing, he ensures that the information in each head is not lost at death but can be communicated to thousands of others and reproduced by them. This is the capacity that puts mankind in a different category than all other creatures, for he can store information additional to that in the gene pool. It is a method that has certainly provided him with great biological advantages. He can live in great numbers in all parts of the earth and possibly eventually in other parts of the universe. It remains to be seen whether there are also dangers in this attempt to preserve information without the shuffling that has been the safeguard of the genetic information for so long. It may be that the very fact of *not* shuffling provides a dangerous conservatism. Perhaps we find it difficult to adjust to the dangers we see, for the very reason that we are so thoroughly well instructed by the past. This may seem paradoxical, but two possibly useful lessons emerge from this approach to memory. One is that the steady state is not maintained by operating according to a fixed set of rules. What the rules do is to provide a system with a memory or repertory of possible actions that have proved effective in the past. It is sensitive to its surroundings and communicates information about them to the memory so that effective items are chosen from those possible. The second lesson is that, though all terrestrial life as we know it uses common materials and principles, yet detailed rules of operation vary enormously and continually. The paradox of life is that constancy and stability depend upon diversity and change, and that these are ensured in the last instance only by death and renewal.

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VI. Gravitation and Relativity

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GRAVITATION IS OBVIOUSLY of great interest to the Air Force. One might even say that it is of central interest, for without it the problems faced would be radically different from what they are. But physicists do not study gravitation in the hope of inventing an anti-gravity device. Nor are they motivated by the desire to make life more comfortable for those who will occupy the Manned Orbiting Laboratory—the use of centrifugal force as a gravity substitute was understood long ago by Newton. Rather, scientists of all kinds, in and out of the Air Force, have three principal objectives. First and foremost, they attempt to improve human understanding of the natural world. Second, they often find in the process that technology is advanced as a by-product. Finally, they help make new knowledge available to those directly engaged in applied science and engineering—through their students, through discussions of all kinds, and through seminars such as this. The accomplishments in the last two categories more than justify the support that basic science has received from the mission-oriented agencies.

SPECIAL AND GENERAL RELATIVITY

The word relativity has two distinct meanings in physics. The special theory of relativity is the successor theory to Newtonian mechanics, and must be used when relative velocities are comparable with the velocity of light. The general theory of relativity is the successor theory to Newtonian gravitation, and must be used when

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gravitational fields are strong. Both theories are the work of Einstein, the special theory having been published in 1905,[1] and the general theory in 1916.[2]

In each case, the extent to which the predictions of the newer theory differ from those of the older can be measured by a dimensionless parameter. In special relativity, the corrections to Newtonian mechanics are of order $(\triangle v/c)^2$, where $\triangle v$ is the difference in velocity between the experimenter and the object that is being observed, and c is the speed of light. In general relativity, the corrections to Newtonian gravitation are of order $(\triangle \phi/c^2)$, where $\triangle \phi$ is the difference in gravitational potential between the positions of experimenter and object, or between different positions of the object during its motion. For example, in measurements of the gravitational red shift in the earth's field, $\triangle \phi$ is equal to the product of the local acceleration of gravity g and the difference in height h of the points between which the red shift is measured. Thus the fractional change in wavelength or frequency is expected to be of order gh/c^2; it actually turns out to be just equal to this. As another example, the motions of planets or light rays near the sun, or of satellites near the earth, depend on the difference in potential between the position of the object and infinity. This $\triangle \phi$ is equal to GM/r, where G is the Newtonian gravitational constant, M is the mass of the sun or the earth, as the case may be, and r is the distance from its center to the object. Thus general relativistic corrections are expected to be of order GM/c^2r.

The magnitudes of these dimensionless parameters in typical cases point up the great difficulty in providing experimental evidence for general, as compared with special relativity. There was a time, thirty or forty years ago, when it was impossible to perform experiments in which $(\triangle v/c)^2$ was comparable with unity, and it was difficult at that time to obtain quantitative verification of the predictions of the special theory of relativity. Those days have long since passed, and there are now many detailed confirmations of the theory. Indeed, none of the many existing high energy particle accelerators would operate as designed if special relativity were not valid, nor would the kinematics of the experiments performed with these accelerators be intelligible. Moreover, the union of electrodynamics, special relativity, and quantum mechanics has yielded a number of results that are in agreement with very precise experi-

ments, and no result that is in disagreement. Thus there can be no reasonable doubt at this time as to the correctness of special relativity within its domain of validity.

While it is now an everyday matter to get $(\Delta v/c)^2 \sim 1$, the general relativity parameter is exceedingly small, and nothing can be done to make it larger. With $h = 10$ meters, $gh/c^2 \sim 10^{-15}$, and at the surface of the earth, $GM/c^2r \sim 10^{-9}$. At the surface of the sun $GM/c^2r \sim 10^{-6}$, and at the orbit of the planet Mercury, this parameter has decreased to about 10^{-8}. Thus *any* experiment that is directed toward confirming or disproving general relativity theory must be arranged so as to measure very small effects with very great precision. It is largely because experiments of this kind push beyond the limits of existing technology that research on general relativity is contributing to advances in applied science and engineering.

It has sometimes been argued that experiments involving large accelerations, such as are produced in centrifuges, can provide evidence on the validity of general relativity theory. In actuality, however, all such experiments can be discussed correctly and completely on the basis of special relativity theory, since permanent gravitational fields produced by massive objects are not involved. The most cogent demonstration of this has been given by Sherwin.[3] He showed that the enormous accelerations experienced by atomic nuclei that undergo thermal oscillations in solids, which are of order $10^{10}g$, do not alter the agreement between observation and the theoretical predictions of special relativity.

THE "CLASSICAL" TESTS OF GENERAL RELATIVITY

In his original paper on general relativity, Einstein[2] proposed three experimental verifications of the theory. These "classical" tests, as they are now often called, consist of measurements of three phenomena: the gravitational shift to longer wavelengths or lower frequencies (red shift) of light in going from a stronger to a weaker gravitational field; the deflection of starlight that passes through the strong gravitational field close to the surface of the sun; and the slow rotation of the major axis, or precession of the perihelion, of the elliptical orbit of an inner planet, Mercury in particular. Let us consider each of these in turn.

The gravitational red shift was first observed in the spectra of

the sun and two white dwarf stars, Sirius B and 40 Eridani B. How-
ever the only precise measurements have been performed in terres-
trial laboratories, where nuclear Mössbauer radiation has been used
over vertical distances of as much as 74 feet.[4] The theoretical pre-
diction that the radiation should decrease in frequency by the frac-
tional amount $\triangle \nu/\nu = gh/c^2$ in travelling upward a distance h has
now been verified with an accuracy of about one percent. This
prediction was first made by Einstein prior to his development of
general relativity,[5] and is not really a test of that theory. Rather it
is a confirmation of what is called the equivalence principle. This
states that observations made in a laboratory that is at rest in a
uniform gravitational field of strength g are in complete agreement
with observations made in a laboratory when it is away from gravi-
tational fields and is subjected to a constant acceleration g. There is
very strong evidence of other kinds for the precise validity of the
equivalence principle.

The theoretical prediction for the red shift can then be derived
by considering the two laboratories illustrated in Fig. 1. The one
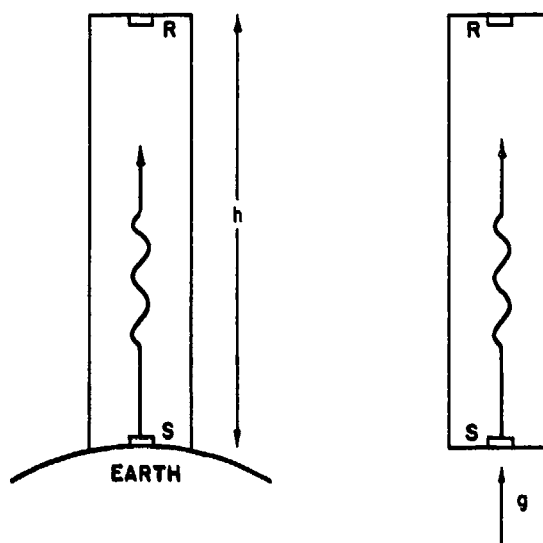
FIGURE 1. Two laboratories in which identical results should be obtained,
in accordance with the equivalence principle. On the left is the actual
laboratory on the earth, and on the right is a hypothetical laboratory that
is away from gravitational fields and has the acceleration g.

on the left is at rest on the surface of the earth, and that on the right is away from the earth but accelerating upwards. According to the equivalence principle, experiments should yield the same results in the two situations. Thus we can calculate the change in frequency of the radiation emitted by the source S and detected at the receiver R by using the arrangement on the right. Since no gravitational fields are involved here, general relativity need not be employed. The radiation requires a time $t = h/c$ to make the upward trip, and during this time the accelerating laboratory increases its upward speed by $\triangle v = gt = gh/c$. Thus by the time R receives the radiation, it is receding with speed $\triangle v$ from S when it emitted the radiation. It thus finds that the frequency is decreased by the amount $\triangle v$ because of the Doppler effect, where $\triangle v/v = \triangle v/c = gh/c^2$.

We see then that a test of general relativity must go beyond the gravitational red shift. The bending of light rays that pass through the strong gravitational field near the sun provides another test of the theory. Here the evidence to date has been obtained during total eclipses, when stars are visible fairly close to the limb of the sun. These observations are not in disagreement with the theory, but are uncertain by roughly 20 to 30 percent, and so cannot be regarded as strong support for it. The prediction is that the light ray should be bent towards the sun, as though it were attracted to it, and that the deflection angle should be $4Gm/c^2r$ radians, where r is the distance of closest approach of the light ray to the center of the sun. This is equal to $1.75(R/r)$ seconds of arc, where R is the solar radius.

There is an interesting and instructive way of viewing the situation described in the last paragraph. Normally one thinks of the motions of light rays and material objects as occurring with respect to an inertial coordinate system which is neither accelerating nor rotating. In such a coordinate system, light rays move in straight lines with constant speed. Now let us suppose that the gravitational field produced by, say, the sun, alters this situation slightly by imposing a new direction in space on the otherwise isotropic inertial system. The word "slightly" is important, because as the light ray moves past the sun, the direction of the gravitation field through which it is passing alters, and if the ray were to maintain a constant angle with the field lines, it would, of course, go in a strongly

curved path around the sun. We assume as part of our qualitative picture that the inertial coordinate system is only very weakly coupled to the local direction of the gravitational field, so that the light ray *tends* to keep the same angle with the field but fails almost completely to do so. The very small parameter GM/c^2r is a measure of its success in following the field lines. The net result is then that the ray moves in nearly a straight line, but is bent *toward* the sun through an angle that is of order GM/c^2r. It should be emphasized that this picture merely provides a way of thinking about the situation once a careful calculation has told us what the answer really is. However, it can also be of some heuristic value in thinking about possible new experiments. We shall return below to its applicability to other situations.

The third of the "classical" tests of general relativity is the observation of the slow precession of the perihelion, or point of closest approach to the sun, of the orbits of the inner planets. Newtonian theory predicts that each planetary orbit is a closed elipse with the sun at one focus. However, perturbations caused by other planets, which can be calculated with Newtonian theory, cause these ellipses to rotate slowly. The orbit of Mercury, which is closest to the sun and hence the best candidate for a test of general relativity theory, is affected most by Venus (since it is closest) and Jupiter (since it is most massive). These perturbations can be calculated quite reliably; when this is done, it is found that about a tenth of the observed precession cannot be accounted for in this way. General relativity predicts that there should be a precession of $6\pi GM/c^2r$ radians during each orbital revolution of 88 days, where r is the mean radius of the orbit. This comes to about 43 seconds of arc per century. In spite of its smallness, this angle has been measured with an accuracy of about one percent, and agrees with the theoretical prediction. We shall return later to some recent observations which cast doubt on this agreement.

The same qualitative picture that was used in discussing the deflection of starlight can profitably be employed here. We imagine that, as Mercury goes about the sun, it attempts to maintain an elliptical orbit with respect to a coordinate system that is weakly coupled to the changing direction of the gravitational field lines. Again, the measure of its success in adapting its orbit to this continually changing direction is the parameter GM/c^2r. This tells

us then that the ellipse rotates slowly in the *same* direction as the planet rotates in its orbit, and that the angle of rotation per revolution is of order GM/c^2r.

THE METRIC TENSOR

Any more quantitative discussion of the extent to which the observations confirm or disprove general relativity must bring in the idea of a metric. Einstein's theory of gravitation is geometrical in nature, and relies on the idea that space and time measurements are distorted by the presence of a massive body. This distortion, or space-time curvature as it is often called, causes corresponding changes in the familiar paths followed by light rays or planets. For example, we normally think of a light ray as travelling in a straight line in the sense that it takes the shortest path from one point to another. But in curved space-time, the shortest path is generally not a straight line, so that light rays are deflected when they pass close to the sun.

The metric tensor, or simply the metric, is a quantitative measure of this curvature. It is easily illustrated in two-dimensional space by comparing the geometry of a plane with that of the surface of a sphere. In a plane, two points that have x coordinates that differ by dx, and y coordinates that differ by dy, are separated by a distance $d\sigma$, where the expression $d\sigma = dx^2 + dy^2$ is called the metric that corresponds to a plane. However we do not have to use the rectangular coordinates x,y; in polar coordinates, where ρ is the radial and ϕ the angular coordinate, the metric is $d\sigma^2 = d\rho^2 + \rho^2 d\phi^2$. This change of coordinate system and metric does not change the nature of the geometry, which is still the flat geometry characteristic of a plane. Suppose now that we replace this last expression by $d\sigma^2 = d\rho^2 + \sin^2\rho \, d\phi^2$, which is practically the same as the earlier expression when ρ is very small. This changes the geometry from flat to curved, since $d\sigma$ is now the distance between two points measured on the surface of a sphere of unit radius. To see this, we need merely note that on a sphere of radius R, $d\sigma^2 = R^2 d\theta^2 + R^2\sin^2\theta \, d\phi^2$, where θ, ϕ are polar coordinates. Then setting $R = 1$ and $\theta = \rho$ gives us our earlier expression. It is always possible to tell from the form of the metric whether the geometry is flat or curved.

It follows then that the concept of curvature can be expressed quantitatively by means of a metric. In flat space-time, that is, in the absence of gravitating bodies, the metric has the form $ds^2 = dt^2 - (1/c^2)(dx^2 + dy^2 + dz^2)$. The path of a light ray is described by the equation $ds = 0$, which means that $d\sigma/dt = c$, where now $d\sigma^2 = dx^2 + dy^2 + dz^2$. We see then that such a path is a straight line and that the speed is constant and equal to c. In order to take curvature into account, we must alter the coefficients of dt^2 and $d\sigma^2$ from unity. Since the parameter GM/c^2r is always small in practical cases, it is natural to express these coefficients as power series in this parameter. We thus arrive at a general form for the metric,

$$ds^2 = dt^2 [1 - 2\alpha(GM/c^2r) + 2\beta(GM/c^2r)^2 + ...]$$
$$-(1/c^2)(dx^2 + dy^2 + dz^2)[1 + 2\gamma(GM/c^2r) + ...]$$

where the coefficients of dt^2, dx^2, etc. are the components of the metric tensor.

The two series have been written in this way in order to facilitate comparison with Einstein's theory, which predicts that the numbers α, β, γ, ... are all equal to unity. Thus we can determine the values of these numbers from the observations, and then express any possible discrepancy from the general relativity prediction in terms of deviations of these numbers from unity. We note first, however, that we can choose the number α equal to unity without any loss of generality, since any other value would simply correspond to a different choice for the value of G; it is really the magnitude of the product αG that determines the Newtonian elliptical orbits.

For the motion of a light ray, we have already seen that dt is equal to $d\sigma/c$ in the absence of a massive body, so that the two quantities are approximately equal even when gravitation is present. Thus we expect that the α and γ terms both contribute to the deflection of light in lowest order, and that the β term will be of higher order. A careful calculation shows that the deflection of light is proportional to the combination $\alpha + \gamma$, or $1 + \gamma$ since we have chosen $\alpha = 1$. The perihelion precession of a planet is a correction to the Newtonian orbit, which is determined by the α term. In this case, dt and $d\sigma/c$ are not of the same order of magnitude. To see this, we consider a circular orbit of a planet of mass m, for which the centrifugal force mv^2/r is equal to the gravitational attraction GmM/r^2, where r is the radius of the orbit. In this case $v^2 = GM/r$, and this will be true in order of magnitude even if the orbit is not

circular. Then the ratio of the β term to the γ term is $[2\beta dt^2(GM/c^2r)^2]/[(2\gamma d\sigma^2/c^2)(GM/c^2r)]=(\beta dt^2/\gamma d\sigma^2)(GM/r)$, which is roughly equal to β/γ since $dt^2/d\sigma^2$ is equal to $1/v^2$. Since this ratio is of order unity, the correction to the Newtonian orbit of a planet involves the β term as well as the γ term. It turns out that the perihelion precession is proportional to $2(1 + \gamma) - \beta$.

It follows that the second of the "classical tests" of general relativity should determine the parameter γ, and the third would then determine β. At present, however, the determination of the deflection of light is sufficiently imprecise so that γ is not well determined at all; it is only the combination $2(1 + \gamma) - \beta$ that is fixed by the planetary orbit precession.

OBLATENESS OF THE SUN

In a recent publication,[6] Dicke and Goldenberg reported their observation that the sun is slightly oblate: the equatorial diameter exceeds the polar diameter by the fractional amount $(5 \pm 0.7) \times 10^{-5}$. They interpret this observation as an oblateness of the mass distribution of the sun as well as of the distribution of visible brightness. It should be noted that if the sun were to rotate with a constant angular velocity equal to the average of that observed on different parts of the surface (period of about 25 days), centrifugal effects would produce an oblateness less than one-quarter of the above value. They therefore assume that the oblateness is caused by a core that rotates very rapidly, with a period of a day or two.

The equatorial bulge in the mass distribution that is caused by the rapidly rotating core affects the orbits of the planets in an entirely Newtonian way; the relativistic effects of the bulge are completely negligible. The Newtonian gravitational force law deviates slightly from the inverse square form that is characteristic of a spherical mass distribution, and causes a precession of the perihelion of Mercury's orbit of 3.4 seconds of arc per century. The remainder of the observed 43 seconds is presumably of relativistic origin, but is evidently less than the Einstein prediction, which as noted above is also 43 seconds. On this interpretation, the Einstein theory is incorrect, or at best incomplete. An alternate theory, proposed several years ago by Brans and Dicke[7] as a development of an earlier theory of Jordan, assumes that gravitation is described not only by

the metric tensor discussed above, but also by a scalar field. This tensor-scalar theory contains an arbitrary parameter, which is essentially the ratio of the magnitude of the tensor to the scalar contribution. This parameter can be chosen so as to reduce the relativistic perihelion precession by the 8 percent suggested by the equatorial bulge. The deflection of starlight should then be about 6 percent less than the Einstein value, but as noted above this is beyond the present precision of the observations.

The foregoing interpretation of the oblateness of the visible sun in terms of a corresponding oblateness of the mass distribution has been questioned in several recent papers. Roxburgh[8] has suggested that the differential Coriolis force associated with the somewhat greater angular velocity of the visible surface of the sun at the equator as compared with the poles, could produce an apparent equatorial bulge equal to that observed without a significant departure of the mass distribution from spherical symmetry. Goldreich and Schubert[9] have argued that the very great differential rotation rate between the core and the surface of the sun, which was postulated by Dicke and Goldenberg, is unstable and would lead to complete mixing in a very short time. Most recently, Sturrock and Gilvarry[10] have remarked that the solar magnetic field could play an important part in determining the visible shape of the sun, so that there may be a substantial difference between the apparent equatorial bulge and the mass distribution.

At the present time we cannot conclude that the observed solar oblateness invalidates general relativity theory. On the contrary, in view of the arbitrariness of the tensor-scalar ratio parameter in the Brans-Dicke theory and the difficulties inherent in Dicke and Goldenberg's interpretation of their observations, it seems most reasonable to assume for the present that the Einstein theory is correct. We discuss below other lines of experimentation which will in time provide independent evidence on the validity of general relativity theory.

OTHER TESTS OF GENERAL RELATIVITY

The paucity of experimental evidence on the validity of general relativity has led to several suggestions for new experiments, all of which are now being implemented. Two of these, proposed in 1960,

involve the generation and detection of gravitational waves,[11] and the measurement of the precession of the spin axis of an orbiting gyroscope.[12] The first of these has produced no definitive results as yet; the second, also in progress, will be discussed in more detail in the next two sections.

More recently, it has been pointed out that the transit times of radar signals that pass through the strong gravitational field near the sun will be affected by general relativity.[13] It is proposed that the time interval between emission of a pulse from the earth and reception of the reflected pulse from Venus or Mercury be measured as the planet passes on the opposite side of the sun from the earth. Again, no results are yet available. A modification of this experiment, on which work has yet to be started, is intended to circumvent the problems associated with uncertainties in planetary radii and topography. It would consist in substituting for the planet as a radar target, a space vehicle that contains a radar transponder and is placed in orbit about the planet.[14]

PRECESSION OF THE SPIN AXIS OF A GYROSCOPE

In Newtonian theory, the spin axis of a torque-free spherical gyroscope remains fixed in space. All of the qualifying adjectives in the preceding sentence are important: Newtonian, torque-free, and spherical. The gyroscope will, of course, precess if it is subject to a torque. It will also precess if it is in an inhomogeneous gravitational field unless it is spherical. Suppose, for example, that the gyroscope has an equatorial bulge and is oriented with its spin axis at 45° to the vertical in the earth's gravitational field. Because of the divergence of the gravitational field lines of the earth and the consequent weakening of gravitational pull with increasing altitude, there will be a stronger downward force on the side of the gyroscope equator that is momentarily lower than the opposite side. This will cause a torque to be exerted on the gyroscope in such a sense as to tend to increase the angle between the spin axis and the vertical. Such a gravity-gradient torque will, of course, make the spin axis precess. However it can be shown that no matter how inhomogeneous the earth's field may be, for example owing to the presence of mountains, there is no torque if the gyroscope is accurately spherical.

Even for a torque-free spherical gyroscope, precession is absent

only in Newtonian theory. Both special and general relativity produce changes in the direction of the spin axis. The special relativity effect is called the Thomas precession, and was first discovered in connection with atomic systems. However, since special relativity is well established in other ways, our interest centers on the general relativity effect, which consists of two parts. The larger part, called the geodetic precession, is proportional to the amount by which the gyroscope moves through the gravitational field of the earth, and is absent if the gyroscope is stationary. This does not, however, mean that there is no geodetic precession if the gyroscope is at rest on the surface of the earth, since the daily rotation of the earth carries the gyroscope with it through the earth's field. In this case the geodetic precession amounts to about 0.4 seconds of arc per year. The effect can be greatly increased by placing the gyroscope in a satellite, since at moderate altitude the satellite will go around the earth some sixteen times a day; the geodetic precession is then about 7 seconds of arc per year.

The smaller but more interesting general relativity effect is called the motional precession. It arises from the fact that the earth is in daily rotation so that the mass of the earth is in continual motion. The Newtonian gravitational field produced by the earth is the same whether the earth is at rest or in rotation. But the Einstein theory predicts a significant difference between the two. An instructive analogy may be drawn between this situation and a somewhat similar electromagnetic arrangement. Suppose that the earth is replaced by a dielectric sphere that has positive electric charge distributed through its volume, and the gyroscope is replaced by a small bar magnet. If the sphere and the magnet are both at rest, no torque is exerted on the magnet. If the sphere is at rest but the magnet moves around it, then the electric field produced by the sphere will appear to the moving magnet to be associated with a small magnetic field, and this will exert a torque on the magnet. This is analogous to the geodetic precession in the gravitational case. Finally, if the sphere is rotating on its axis, the circulating electric current of the moving charges will produce an external magnetic field, and this will exert a torque on the magnet whether or not the magnet is at rest. This is analogous to the motional precession in the gravitational case.

General relativity predicts motional precession whether or not

the gyroscope is moving. Near the surface of the earth it is of the order of a tenth of a second of arc per year. The reason it is of great interest in spite of its smallness is that it is the *only* manifestation now known of the influence of mass motion on gravitation. This is the reason that very great effort is being directed toward the highest possible precision in measuring the gyroscope precession.

A detailed calculation shows that the angular momentum vector **S** of the gyroscope changes with time in the following way:[12]

$$\frac{d\mathbf{S}}{dt} = \Omega \times \mathbf{S}$$

$$\Omega = \frac{\mathbf{F} \times \mathbf{v}}{2mc^2} + \frac{3GM}{2c^2r^3}(\mathbf{r} \times \mathbf{v}) + \frac{GI}{c^2r^3}\left[\frac{3\mathbf{r}(\omega \cdot \mathbf{r})}{r^2} - \omega\right]$$

The first equation states that the change in **S** is at right angles to **S** itself; thus **S** does not change in magnitude, but precesses about the direction of the vector Ω at a rate equal to the magnitude of Ω in radians per second. The second equation is an expression for the precession angular velocity Ω. The first term is the Thomas precession, and depends on the nongravitational force **F** exerted on the gyroscope, and on the gyroscope's mass m and velocity **v**. Since this term does not involve the mass M of the earth or the Newtonian gravitational constant G, it cannot arise from general relativity, and as remarked above is of special relativistic origin. The second term is the geodetic precession, and depends both on **v** and on the vector position **r** of the gyroscope with respect to the center of the earth. The third term is the motional precession, and involves the moment of inertia I and the angular velocity vector ω of the earth. It evidently arises from rotation of the earth, and is present whether or not the gyroscope is in motion since it is independent of **v**.

The experiment now under way is described in the next section, and will be performed in a satellite. The main reason for choosing an orbiting rather than a laboratory gyroscope is that the former is in free fall and hence does not have to be supported against gravity. This means that the inevitable small misalignment of center of mass and center of support of the gyroscope does not exert a torque which could confuse the observations if it were present. A secondary reason is of course that the geodetic precession is much larger in orbit than on the surface of the earth; on the other hand, the

motional precession is roughly the same in the two cases. Further, the relatively uninteresting Thomas precession is zero in orbit, since $\mathbf{F} = 0$ there.

Both the geodetic and motional precessions can be viewed in terms of the qualitative picture described earlier in this paper. We imagine that as the gyroscope goes about the earth, it attempts to maintain a fixed direction for its spin axis with respect to a coordinate system that is weakly coupled to the changing direction of the gravitational field lines. The measure of its success in adapting its spin axis to this continually changing direction is the parameter GM/c^2r. This tells us that the spin axis rotates slowly in the same direction as the gyroscope rotates in its orbit, and that the angle of rotation per revolution is of order GM/c^2r. Actually, the geodetic (second) term in the expression for Ω shows that for a circular orbit of radius r, $\mathbf{r} \times \mathbf{v}$ integrates to $2\pi r^2$ over one revolution, so that the geodetic precession is $3\pi GM/c^2r$ radians per revolution.

The same picture can be applied to the motional precession. Imagine first a gyroscope that is fixed over one of the poles. As the earth rotates, it tends to "drag" its gravitational field with it, and this tends to make the spin axis of the gyroscope rotate in the same direction as ω. In this situation, $\mathbf{r}$ is parallel to ω and the square bracket term in Ω is equal to 2ω, in agreement with the picture. On the other hand, if the gyroscope is fixed over the equator, $\mathbf{r}$ is perpendicular to ω, and the square bracket term is equal to $-\omega$. Here the qualitative picture tells us that the earth's field is getting weaker as it extends out from the earth, so that the dragging effect is more pronounced on the side of the gyroscope toward the earth than it is on the side away from the earth. Thus the gyroscope spin axis tends to rotate in the opposite direction from the earth, in agreement with the calculation.

The motional precession cannot be expressed in terms of the parameters β and γ introduced earlier, since the metric tensor needed to account for earth rotation is more complicated than that used before for the nonrotating earth or sun. However the geodetic precession can be calculated in this way, and it turns out that the number 3 in the numerator of the second term of Ω is replaced by $1 + 2\gamma$. Thus measurement of the geodetic precession is somewhat more sensitive to the value of γ than measurement of the deflection of starlight.

THE GYROSCOPE EXPERIMENT

The gyroscope experiment is being performed in the Stanford University Physics Department by Fairbank and Everitt.[15] The gyroscope itself will consist of a very homogeneous quartz sphere, $1\frac{1}{2}$ inches in diameter, and coated with a thin layer of niobium. This sphere will be supported in vacuum by electrostatic forces, which will have to overcome only the very small differential between earth gravity and the acceleration of the satellite. The satellite will not be quite in free fall, because of aerodynamic drag, solar radiation pressure, etc., but the difference should not be of greater order of magnitude than 10^{-7}g. Once in orbit, the gyroscope sphere will be spun up to speed by helium gas jets, and kept cold enough so that the niobium coating is superconducting.

The main reason for operating at liquid helium temperature is that a rotating superconductor possesses a magnetic moment that is accurately aligned with its axis of rotation, and the measurement of this moment therefore makes possible a precise determination of the direction of the gyroscope spin axis. This effect was predicted theoretically by London in 1950,[16] and observed experimentally in 1964.[17] The London moment corresponds to a magnetic field along the spin axis of $10^{-7}\omega$ gauss, where ω is the angular velocity of rotation in radians per second. It will be detected by a superconducting loop that encircles the gyroscope, as indicated in Fig. 2. Changes in the magnetic flux that links this detector loop will be measured by a modulator that is part of the same superconducting circuit, as illustrated schematically in Fig. 3.

Figure 4 is a sketch of the probable configuration of the apparatus, which contains a telescope of 4 inches aperture and four gyroscopes. The telescope will be aligned on a star and provide the reference direction. There will of course be corrections for aberrations that arise from the motion of the satellite around the earth and of the earth around the sun; these are easily calculated, and will provide convenient checks on the performance of the telescope and London moment read-out. A secondary reason for low temperature operation is the extreme constancy and uniformity of the temperature of the entire apparatus, and the consequent freedom from thermal distortion. It is expected that angular measurements with accuracy exceeding 0.01 seconds of arc can be made over the course of a year.
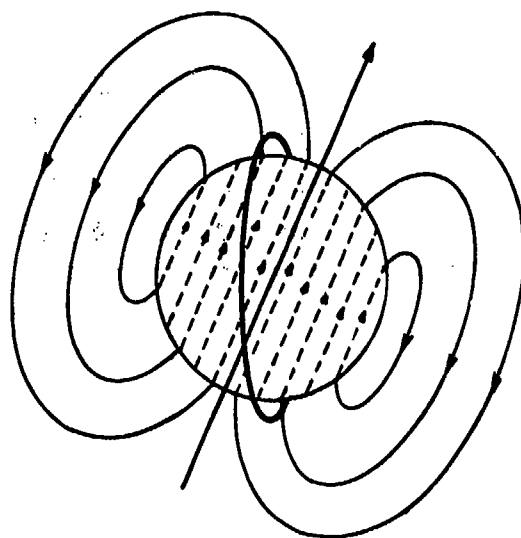
LONDON-MOMENT FIELD H = 10⁻⁷ω GAUSS

FIGURE 2. Principle of the London moment read-out.
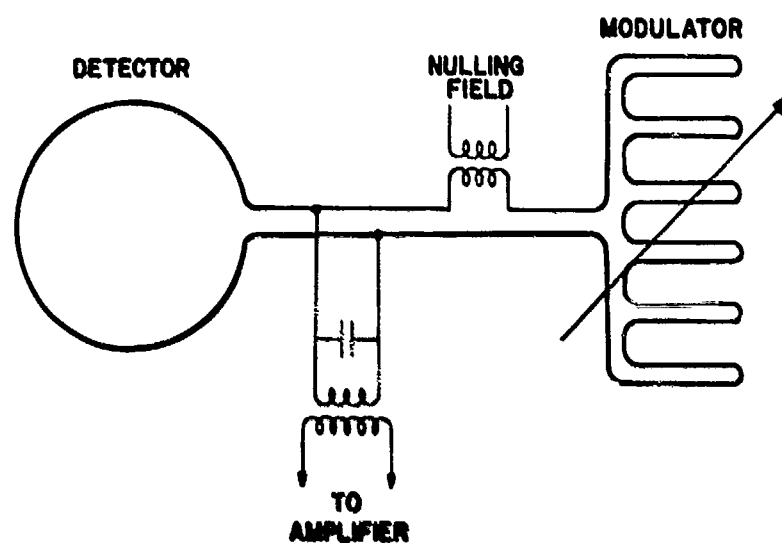
FIGURE 3. Schematic illustration of the superconducting detection circuit. The loop to the left encircles the gyroscope, and the arrow indicates that the inductance of the right-hand part of the superconducting circuit can be varied periodically to provide a signal that is proportional to the flux linked by the detector loop.
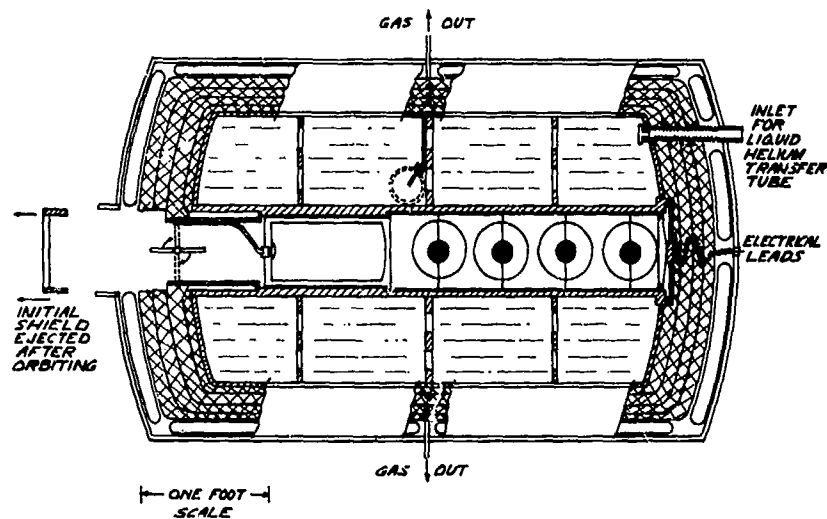
FIGURE 4. Flyable low temperature gyroscope experiment.
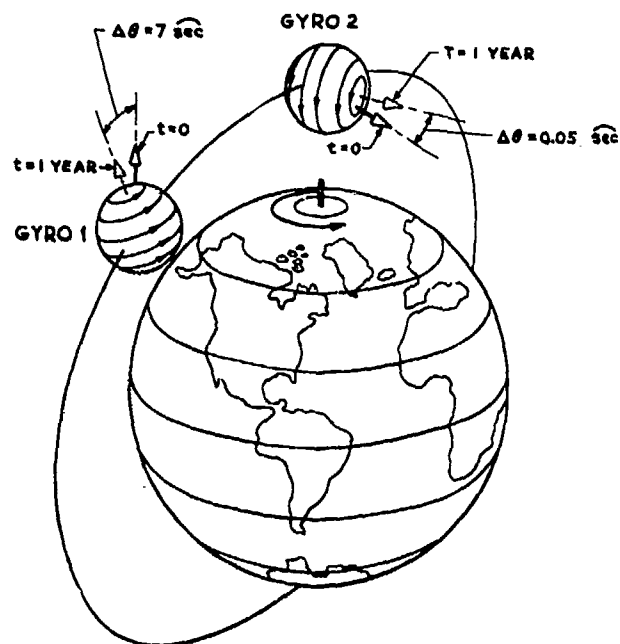
FIGURE 5. Relativisitic precessions of gyroscopes in a 500-mile polar orbit. The two gyros of type 1 will measure the geodetic precession, and the two of type 2 will measure the motional precession that arises from the rotation of the earth.

It is planned to place the apparatus in polar orbit, as shown in Fig. 5. Two of the gyros will have their spin axes in the plane of the orbit and parallel to the earth's rotation axis (gyro 1 in the figure), and two will be oriented perpendicular to the plane of the orbit (gyro 2). The first pair will be sensitive to the geodetic precession alone, and the second pair will be sensitive to the motional precession alone. Reasonable estimates of the rate of loss of liquid helium indicate that readings should be obtainable during most or all of the first year after launch.

As remarked at the beginning, experiments on general relativity push beyond the limits of existing technology. The gyroscope experiment provides an excellent example of progress in low temperature engineering and magnetometry that was stimulated by research in basic science.

ACKNOWLEDGMENT

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VII. Toroidal Plasma Confinement For Fusion

Melvin B. Gottlieb

IT WAS JUST about fifteen years ago that the effort to "tame the hydrogen bomb" started in the United States, the United Kingdom and the Soviet Union. It was, of course, well known that the vast amounts of deuterium in the oceans of the world represented an almost unlimited potential source of energy through the D — D reaction.

$$D + D \nearrow T + p + 4 \text{ MeV}$$
$$\searrow He^3 + n + 3.3 \text{ MeV}$$

The D — T reaction

$$D + T \rightarrow He^4 + n + 17.6 \text{ MeV}$$
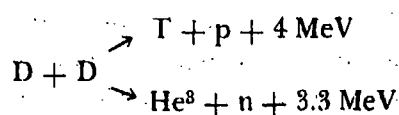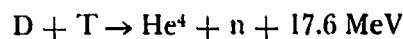
has similar potential—in this case the basic fuels become deuterium and lithium, the latter being used to provide tritium through an (n, T) reaction. The hydrogen bomb shows that this energy may be released explosively—but as yet the feasibility of a controlled thermonuclear reactor remains to be demonstrated.

At the outset it was recognized that in order to achieve a reasonable reaction rate the materials would have to be brought to extremely high temperatures—of the order of $10^{8\circ}$ K; that at such high temperatures the atoms would all be ionized, thus forming a

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plasma; and that the use of magnetic fields offered the possibility of providing insulation between the hot plasma and the cold walls.

I would like to give my own view of the present state of the work on the containment of a hot plasma by a magnetic field.

First, I will describe some charged-particle trajectories in relatively simple magnetic and electric fields, and then show how these considerations affect a few of the many field geometries that have been proposed, in therms of the equilibrium and stability of a plasma confined in a magnetic field.

Let us start with the simple case of a particle moving at right angles to a constant magnetic field. It executes a circle of radius $mv_\perp c/eB$, m, $v_\perp$, and e being the mass, velocity, and charge of the particle, B the field strength, and c the velocity of light. If the initial velocity is not perpendicular to the field, then the motion along the field, $v_\parallel$, is unaffected; the motion perpendicular is the circle as before—the total motion being a spiral around the field lines. If the magnetic field intensity varies slightly about the orbit, then a drift takes place as shown in Fig. 1—a case where the magnetic field is slightly stronger at the top than at the bottom. Note that the + and − particles (ions and electrons) drift in opposite directions. Suppose an electric field is applied perpendicular to a uniform B
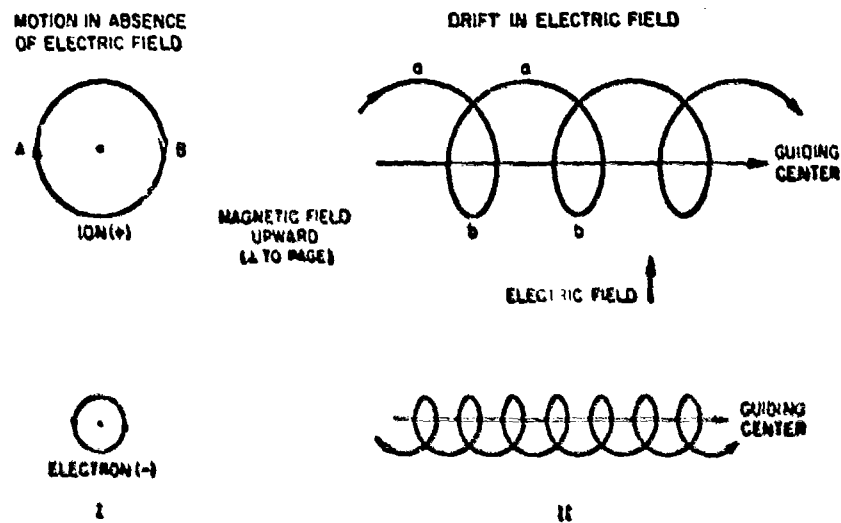
Figure 1. Charged-particle drift in an inhomogeneous magnetic field.

field. Now both kinds of particles drift as shown in Fig. 2, at the same speed cE/B, at right angles to both E and B, and in the $\mathbf{E} \times \mathbf{B}$ direction.

Now let us consider what happens when a collection of ions and electrons is placed in a solenoidal magnetic field as shown in Fig. 3. The particles circling around the field are thus kept off the walls but can escape freely out the ends. To at least partially block the ends, the field intensity can be increased at the ends—forming so-called magnetic mirrors. Now the force exerted by a magnetic field

MOTION IN HOMOGENEOUS
MAGNETIC FIELD

DRIFT IN INHOMOGENEOUS
MAGNETIC FIELD

GUIDING
CENTER

ION (+)

MAGNETIC FIELD
UPWARD
(⊥ TO PAGE)

INCREASING MAGNETIC FIELD ↑

ELECTRON (−)

GUIDING
CENTER
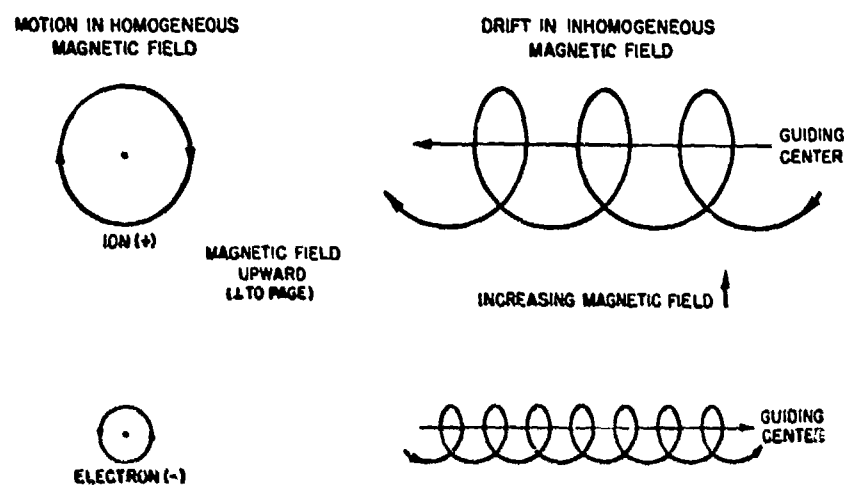
FIGURE 2. Charged-particle drift in crossed electric and magnetic fields.
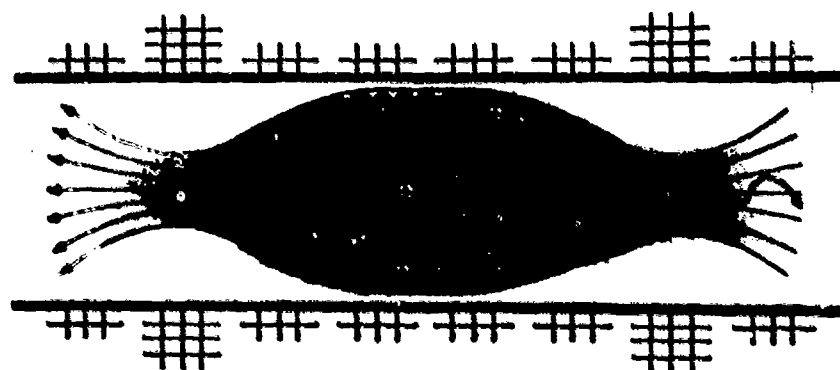
FIGURE 3. Charged-particle motions in a strong magnetic field.

on a moving charged particle is always perpendicular to B. In the region where the lines of force are converging, there is a small component of this force urging the particle toward the weaker field. This force may be large enough to prevent the particle from escaping out the end; a reflection occurs. Of course, a particle moving mainly parallel to B is relatively unaffected. Indeed, it turns out that only particles in the central region, with velocity vectors lying within a cone of generating angle $\theta = \csc^{-1}R^{1/2}$, where R (termed the mirror ratio) is the ratio of the magnetic field magnitude in the mirror to that at the central plane, escape through the mirrors. The others are reflected—they are contained. These particles will be lost only if a collision or other scattering process occurs which changes the velocity vectors to one within the escape cone.

Can these ends be avoided completely by closing the lines in the form of a torus? (See Fig. 4.) The field in a torus falls off as $1/r$, where r is the distance from the toroidal axis. The field is non-uniform and, as shown previously, ions and electrons drift in opposite directions. An electric field, transverse to B, results from this charge separation. Particles are inhibited by the magnetic field from flowing in such a direction as to neutralize this charge accumulation. In this crossed electric and magnetic field situation the plasma drifts rapidly outward in the manner suggested by Fig. 2.

Thus we say that a plasma in a simple toroidal magnetic field does not possess an equilibrium. Let me differentiate this from the matter of stability. To find out whether a confined plasma is stable, we apply a small perturbation and see whether this perturbation grows in amplitude. In the toroidal case just described, no perturbation was applied; still electric fields were developed within the plasma which would drive the plasma outward. For an equilibrium to exist we demand that a steady state exist, exhibiting no growing flows or electric fields. This concept of equilibrium involves a time scale. Certainly a plasma contained by a magnetic field is not in thermodynamic equilibrium, and therefore will not remain indefinitely in the steady state we have envisaged. This point will be amplified later.

Are there magnetic configurations with closed lines—toroids, topologically speaking—that do possess an equilibrium? Yes, there are many such configurations. If, instead of forming closed circles, the lines of force are caused to twist as shown in Fig. 5, they do not

close on themselves (except in certain cases where the rotation per loop around the torus is a rational fraction of 2π). Each line of force, when followed many times around, tends to form a surface—termed a magnetic surface—and the system now consists of an infinite set of nested surfaces. Any local charge excess tends to leak

Section AA'
Field Lines out of Paper

Section BB
Field Lines into Paper
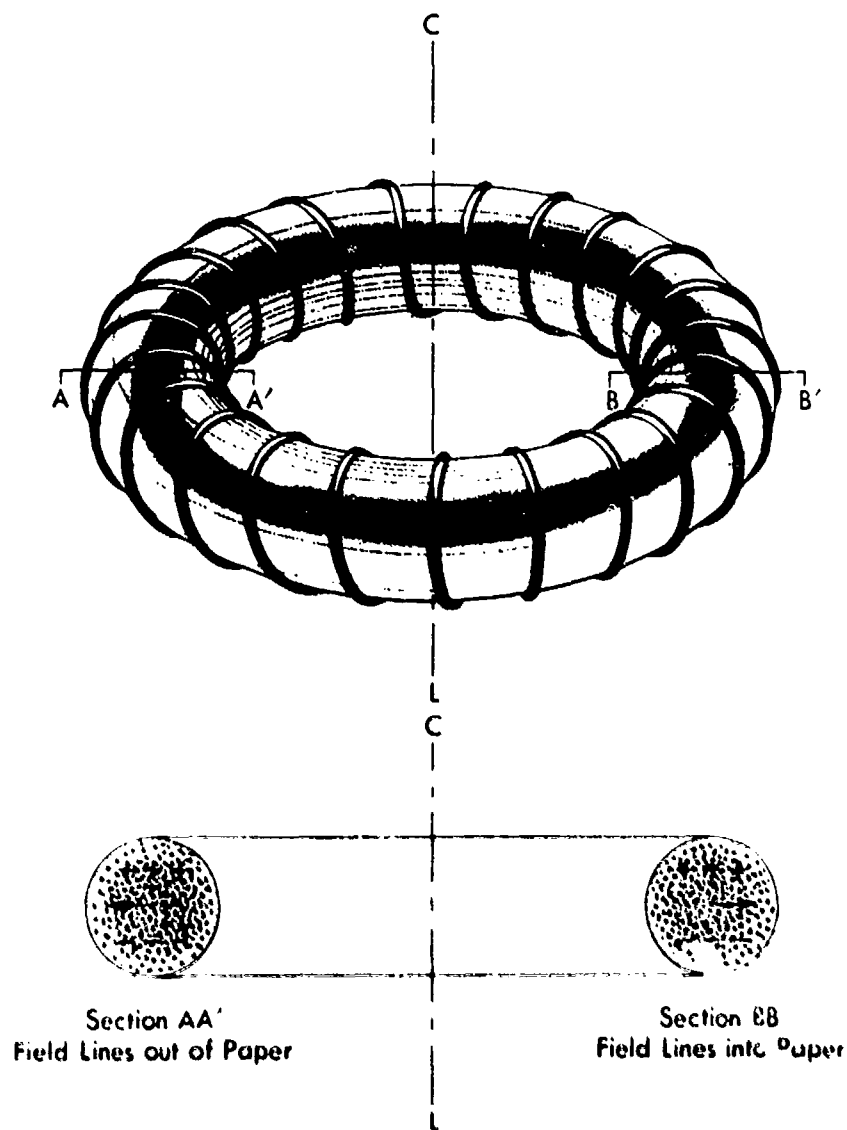
FIGURE 4. Toroidal magnetic field.
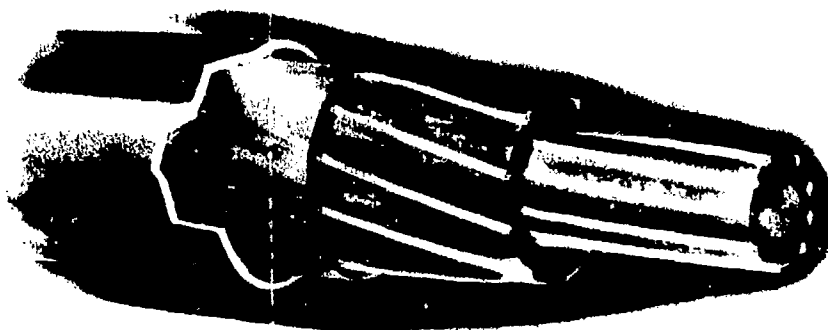
FIGURE 5. Twisted field lines which form magnetic "surfaces."

off along the field lines, distributing itself uniformly. Such a sym-
metric charge distribution causes only an electric field perpendicular
to the wall. The resulting drifts, being (as shown earlier) perpen-
dicular to E and B, are thus parallel to the walls, causing no losses
to the wall.

It is appropriate to define another term which turns out to be
important—shear. When the lines of force in successive surfaces
twist at different rates, we say there is shear in the field.

How do we get magnetic surfaces? Fig. 6 shows a number of
methods. In each case the tube should be regarded as surrounded
by coils providing the magnetic field which goes around the torus
(the long way). Only the additional features causing the twist are
shown. The torus may be deformed (figure-8 stellarator), view (a);
helical windings may be added to the solenoidal winding (helical
stellarator), providing a transverse field component, view (b); a
current induced in the plasma will provide rotation of the field
lines (e.g., Soviet Tokamak), view (c); a current-carrying rod (e.g.,
levitron or spherator), view (d), may be used.

In all these cases except the first-mentioned (figure-8 stellarator)
the magnetic field does possess shear.

Before moving on to describe the experimental results there is
one additional concept which is of importance—namely, contain-
ment time. A successful reactor obviously requires a greater power
output than power input. The plasma particles must be heated
before they will react. Each particle has, on the average, 10 keV of
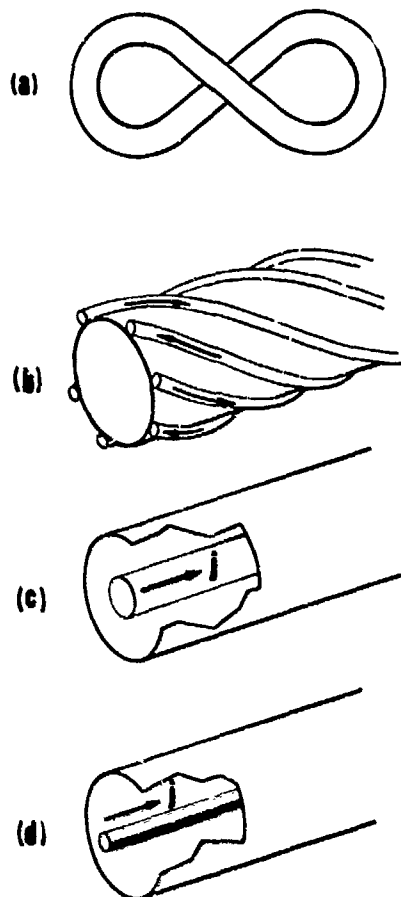
FIGURE 6. Configurations producing magnetic surfaces.

kinetic energy. If it hits the wall, this represents an energy loss. A reaction yields roughly 10^3 times as much energy. To just break even, energywise, at least one particle must engage in a nuclear reaction for every thousand particles that escape. At temperatures of the order of those required for a reactor, the above condition yields the so-called Lawson criterion

$$n\tau > 10^{14},$$

where n is the number of ions per cubic centimeter and τ the time (in seconds) before the average ion escapes to the walls. Considerations of power density and maximum available magnetic field indicate that n would be in the range 10^{14} to 10^{17}. Thus minimum con-

tainment times of the order of .001 to 1 second are required. Now this is a very long time when one considers the fact that the ion thermal speeds are about 5×10^8 cm/sec. This brings us to the crux of the matter. What are the processes which result in the loss of particles? As I stated before, the system is not in thermodynamic equilibrium from a number of standpoints. First, the plasma is thin compared with an absorption length for radiation. Therefore, it does not (fortunately indeed) radiate as a blackbody. Second, we require that the density fall off to a low value at the walls; there is a density gradient. Third, the velocity distribution may be significantly non-Maxwellian.

We are concerned here with particles escaping across the magnetic field lines. Obviously, a single particle circling around the field lines should remain there indefinitely (if it doesn't radiate). When many particles are present, collisions occur. After each collision the particle starts moving in a different circle—displaced from the original one. This pr   results in a gradual diffusion from regions of high density to   regions of low density. The containment time calculated on the basis of this mechanism should be proportional to $B^2 T^{1/2}$ and is long compared with our requirements Much more serious is the matter of losses due to instabilities. Suppose, for example, that a wavelike disturbance exists in the plasma, causing oscillating electric fields as well as varying particle densities. In a region where the field exists, all of the particles or a significant fraction of them will drift together. It is apparent that low-frequency electric fields can be particularly effective in causing large motions.

It happens that a plasma confined by a magnetic field tends to be unstable with respect to various wave-modes, and so our task turns out to be that of finding means of suppressing as many of these modes as possible and of limiting the amplitude of those that do grow. This is a complex task, but, as you will see, we are making substantial progress.

MODEL C STELLARATOR RESULTS

The Model C stellarator was built in order to find the containment properties of a toroidal, sheared magnetic field; a helical stellarator. I do not propose at this time to go into the techniques of plasma creation, impurity control, and measurement. Suffice it to

say that these are complex matters and that a great deal of ingenuity and hard work has been involved. I will simply summarize the results of the group led by Spitzer and Grove. Measurements have been carried out over an extremely wide range of plasma parameters on Model C, far more than on any other plasma device. These are shown in Fig. 7. The results on the critical quantity-containment time are shown on Fig. 8. Within a factor of 2, all the points lie along a line called "Bohm time." The containment time is proportional to B, not B^2, and to $1/T$ instead of $T^{1/2}$. I will come back to "Bohm time" shortly. Suffice it to say, at this point, that until very

METHODS OF HEATING PLASMA

METHODS OF MEASURING

ION TEMPERATURE:

1. DOPPLER BROADENING
2. NEUTRON YIELD
3. VELOCITY DISTRIBUTION OF IONS
4. VELOCITY DISTRIBUTION OF NEUTRALS

ELECTRON TEMPERATURE:

1. CONDUCTIVITY
2. SPECTROSCOPICALLY
3. MICROWAVE ABSORPTION AT $2\omega_{ce}$
4. LANGMUIR PROBES
5. VELOCITY DISTRIBUTION OF ELECTRONS

1/2 (ELECTRON + ION TEMPERATURES):

1. DIAMAGNETIC PRESSURE
2. MASS MOTION
3. SEGMENTED APERTURE LIMITER
4. "FORCE-FREE" EQUILIBRIUM

FIGURE 7. Plasma parameters in the Model C stellarator.
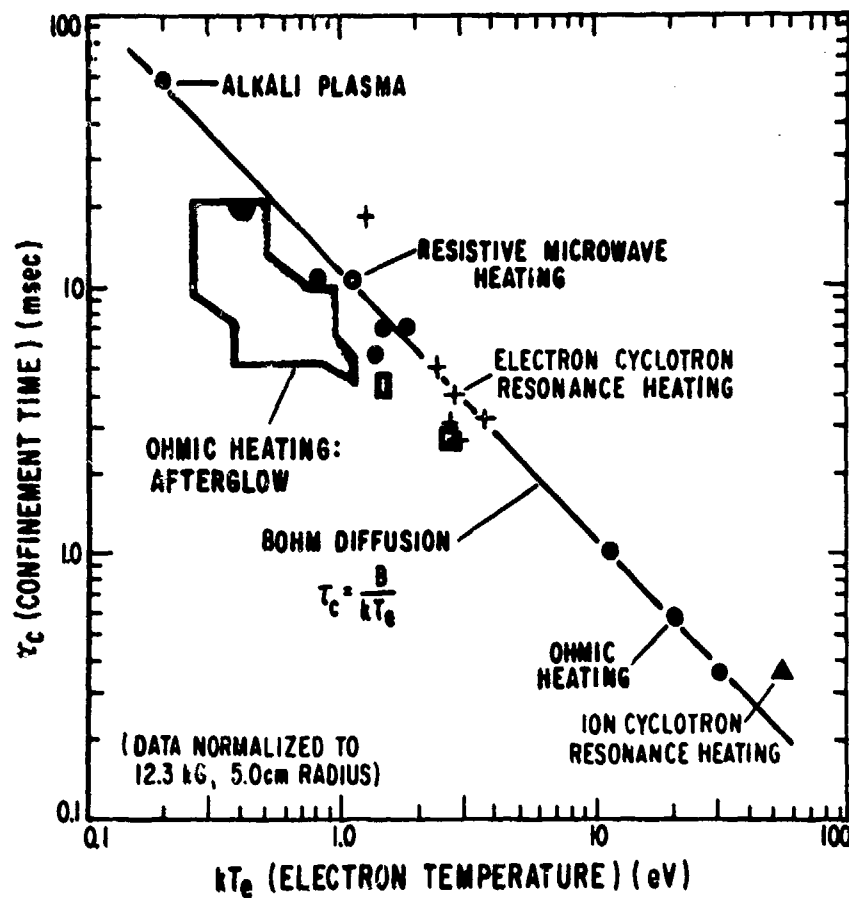
FIGURE 8. Containment time vs magnetic field in the Model C stellarator.

recently there existed no adequate theoretical derivation of the Bohm formula.

When one examines the local behavior of this plasma, large fluctuations are observed in the density; fluctuating electric fields are observed. How do we arrive at some understanding of what is going on here? Well, for a hint, let's go back and look at some of the mirror system results.

In a magnetic mirror configuration the lines of force all bulge out—which is equivalent, from the standpoint of Maxwell's equation, to the statement that the magnitude of the field falls as one moves away from the axis. As I pointed out earlier, in a nonuniform

B field the particles drift—electrons in one direction, ions in the opposite—perpendicular to B and ∇ B. In a cylindrically symmetric plasma, this drift about the axis produces no charge separation. If, however, the surface is perturbed slightly (as shown schematically in Fig. 9), the drifts do produce a charge separation. Furthermore, the resulting electric field produces an additional motion in such a direction as to cause the original bump to grow. Electric charges,

B OUT ∇ B RADIALLY INWARD

E

PLASMA   E
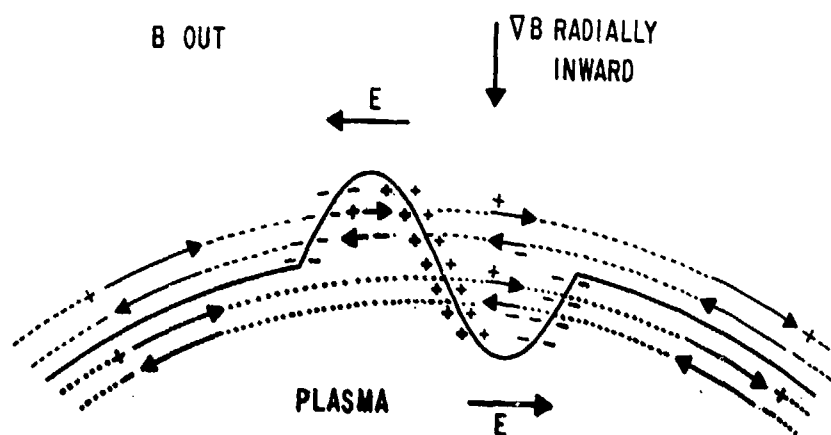
FLUTE INSTABILITY

FIGURE 9. The flute instability.

which may initially grow at a particular local region, tend to spread out along a field line and the "bump" tends to take on the form of a long ripple or flute.

Note one important idea. If the magnetic field gradient were in the opposite direction (if the curvature bulged inward), the ions and electrons would drift in the opposite directions, the charge accumulations would be interchanged ($+$ and $-$), the electric fields reversed, and the bulge (instead of growing) would be pushed back down—the system would be stable against a flute. Ioffe, in 1955, reported experimental results supporting these ideas. He added to the ordinary mirror system a set of parallel bars, each bar carrying a current opposite in direction from its neighbors (Fig. 10). While the pattern of the lines of force is somewhat complicated, it is clear

STABLE | UNSTABLE | STABLE
REGION | REGION | REGION

(A)   (B)

MAGNETIC MIRROR DEVICE WITH IOFFE BARS
(C)
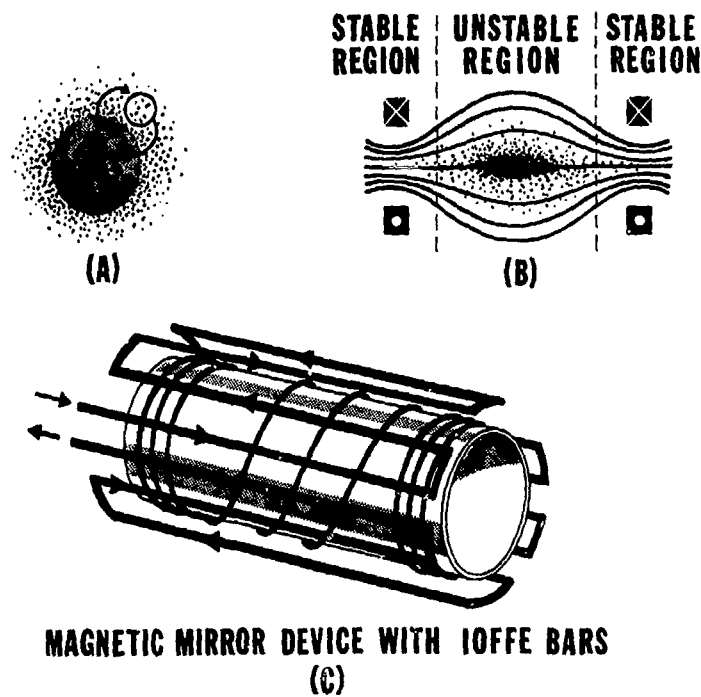
FIGURE 10. Ioffe system for producing a field minimum in the central region.

that at the axis the net field due to the bars is zero, and that as one moves outward toward the bars, the field due to the bars increases. Thus, there exists a region inside such that the magnitude of B increases as one moves outward in any direction—i.e., there is a minimum B region. Ioffe found a factor of 30 increase in confinement time when sufficient current was passed through the bars to create a minimum $|B|$.

Since that time another instability, called a drift mode, has been identified. It also tends to form long flutes. The description of the particle motions is somewhat complicated, and I won't attempt it here. Suffice it to say that this mode is even more dangerous than the mode I described, in that it is a low-frequency mode and harder to stabilize. It tends to exist whenever there is a pressure gradient (and therefore it sometimes is called a "universal" mode). Additional driving terms are electron-ion collisions, field curvature, and plasma

current. According to theory, this mode too should be suppressed by a minimum B field geometry.

However, true minimum B in toroidal form, with the lines of force not crossing a wall, is impossible, for in order to have a minimum B the lines of force must bend outward. This is incompatible with their staying inside a toroidal region.

A possible solution was proposed by Furth and Rosenbluth. The idea may be described as follows. Suppose the lines of force alternately curve outward (tending to be a stable region) and inward (tending to be unstable). Now an instability with a long wavelength may involve both regions, and for such modes it is of interest to investigate the net stabilizing or destabilizing effect—averaged along the line. The pertinent quantity turns out to be $\oint d\ell/B$, where $d\ell$ is an element of length along the line of force. If this quantity decreases as one moves outward toward the wall, then long wavelength drift waves should be stable. This concept is called average minimum B. How about short wavelengths? It turns out that short wavelengths will also be stable provided the regions of "good" and "bad" curvature are sufficiently close together (so-called short connection lengths). This can be viewed essentially in the following terms. A disturbance which starts to grow in the bad region tends to spill plasma over into the good region—the rate of flow being essentially the ion thermal velocity. If the plasma reaches the good region in a time shorter than the growth time of the instability, a strong stabilizing effect should take place. High ion temperatures are therefore advantageous.

Thus an average minimum B, together with short connection length, should tend to stabilize the drift wave.

There is another stabilization scheme which should in theory also be effective, and that is to have strong shear in the magnetic field.

There is a considerable amount of corroborative experimental evidence for both these stabilization methods. I will mention only a few here.

The minimum-average-B, short-connection-length concept has been applied in experiments by Kerst at Wisconsin and Ohkawa at General Atomic. The apparatus consists of a toroidal tube containing four circular current-carrying rods shown schematically in Fig.

11. Typical lines of force are shown in this figure. There is one set of lines which cross at the center (at which point the field must be zero), labelled "Separatrix" (ψ_s). Lines closer to the rods than ψ_s circle only one rod. Lines further from the rods enclose all four rods; for example, that labeled ψ_{crit} (a critical line). The region inside ψ_{crit} should be stable from the $\oint d\ell\, B$ or average minimum B standpoint. The region outside ψ_{crit} should be unstable. This may be anticipated from the fact that as one moves outward the lines of
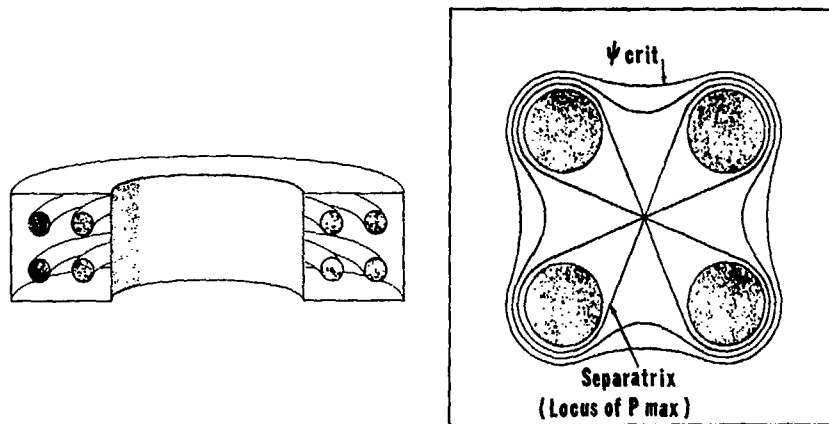
FIGURE 11. The toroidal octupole.

force gradually approach circles. Circles, of course, have only "bad" curvature. Thus far experiments have been carried out only at relatively low densities. The experiments indicate that the plasma outside ψ_{crit} shows large density fluctuations, but inside ψ_{crit} the fluctuations are very small, indicating that the containment time may be very long compared with the Bohm time. Thus far, actual confinement time measurements are limited due to the fact that supports for the four rods extend into the plasma. The observed plasma lifetime is in agreement with that which would be expected due to these obstructions.

Experiments are now being planned in which the rods, instead of being supported mechanically, will be supported by magnetic fields. In order to eliminate current feeds which provide similar obstructions, the rings will be superconducting. Existing technology now seems adequate for such a device.

For evidence on shear stabilization let us look at the results of an experiment by Chen. He has a long, straight solenoid with a uniform magnetic field parallel to the axis. Down the center of the table runs a long rod through which a current may flow. The magnetic field is twisted and sheared by this current. At one end of the tube is a (constant) thermal source of potassium plasma. A cross section of the tube is shown in Fig. 12. A probe, measuring density, is moved along the path shown by the dotted line. One anticipates that, since
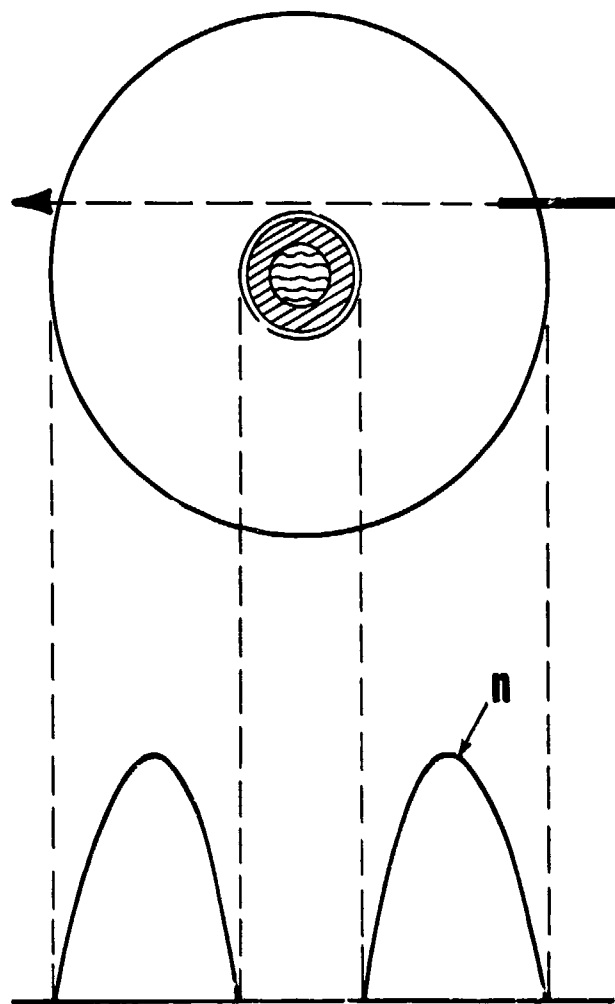
FIGURE 12. Solenoid with shear produced by current in an axial rod.

the plasma is absorbed both at the outer wall and at the rod, the density distribution should be of the form shown. Maintaining a constant plasma source, the shear is increased by bringing up the current in the rod. Figure 13 shows that the density continues to
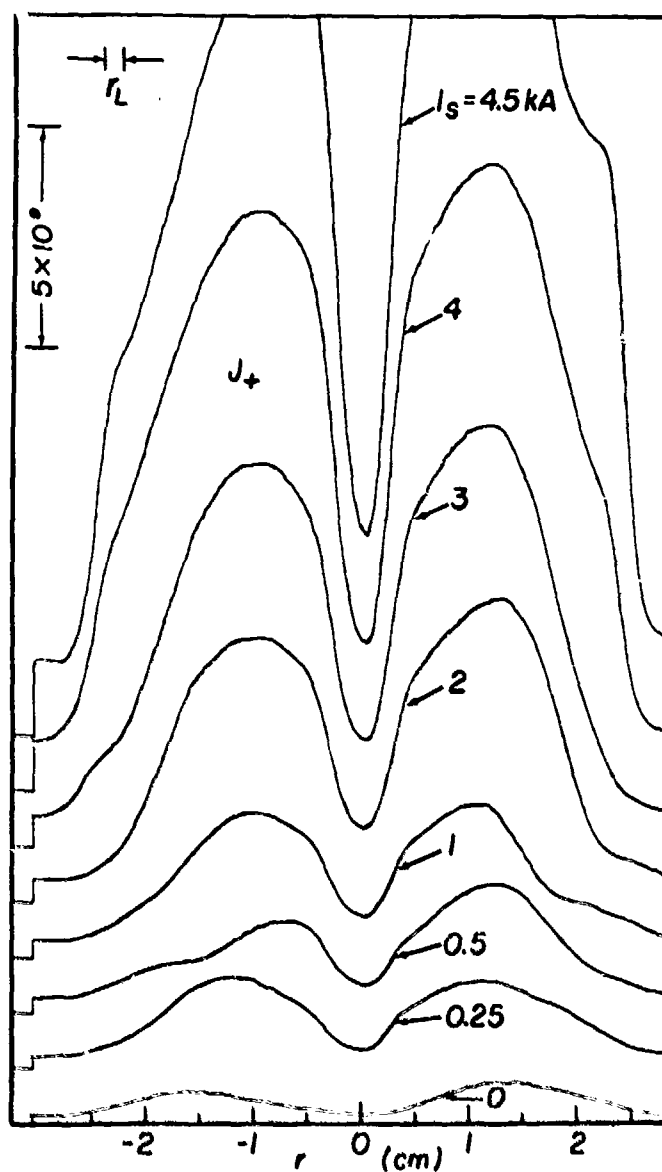
Figure 13. Density as a function of radius as the shear is varied.

rise as the shear is increased, indicating that the losses are indeed decreasing with shear. Noise measurements on this plasma also show the strong stabilizing effects of shear.

There is one other factor which has a stabilizing influence and that is viscosity. Viscosity would have only a very small effect in the regime of interest to CTR; however, it is possible to test the validity of the theory by doing experiments in which viscosity does play a role. Recently Grove calculated that by going to a regime of low density (so that the collisional driving terms are small), and to high ion mass (using xenon so as to increase the Larmor radius and thus increase the viscosity), and by using somewhat more shear than was heretofore available, he could theoretically get into a regime where all but one mode was stable and this one unstable mode was predicted to have a slow growth rate.

At this point I need to go off on a tangent again. The relevant point will emerge shortly. Plasma theory is an enormously complicated field. The basic equations are nonlinear. Perturbation techniques are used to linearize the equations in order to obtain solutions. The descriptions of the wave modes, for example, are valid only so long as the wave amplitudes are small. The growth rates predicted by linearized theory cannot continue to be correct as the amplitude increases. Obviously the energy in the wave must remain finite—there must be a limiting amplitude to which the wave grows. This amplitude should then correspond to a definite plasma loss rate. There exists at this time no completely satisfactory method of calculating the limiting amplitude. Approximate calculations (quasilinear) for some modes, by Galeev, give a diffusion coefficient of the order of $\gamma/k_{\perp}^2$, where γ is the growth rate predicted by linear theory, and $k_{\perp}$ is the wave number of the unstable mode. Finally we come to the point. This theory predicts a diffusion rate that leads to Bohm diffusion for the regime in which Bohm diffusion was observed in the C stellarator. But to go one step further, in the xenon low-density regime referred to earlier, this same theory predicts that the diffusion rate that results from the unstable mode referred to should drop by about an order of magnitude. Surprisingly enough, the experiment performed recently did show a diffusion rate lower by a factor of 5.

In summary, there is beginning to be a very encouraging correspondence between our theoretical ideas and our experimental ob-

servations. I do not for a moment believe that our knowledge is at all complete or even adequate. But we are making solid progress.

There are important developments in other confinement experiments which I have not been able to discuss in this limited time. These include the Astron experiment involving relativistic electron layers, the programs involving high energy ion injection into mirror geometries and the successful creation, without apparent instabilities, of high-β (high density and temperature) plasmas in the theta pinches. Since I personally feel that the hope for a successful future CTR is along the path outlined by the toroidal work I have described, this has been my emphasis.

VIII. The Internal Structure of Shock Waves

HANS W. LIEPMANN

INTRODUCTION

FIGURE 1 is a flow picture of a supersonically flying projectile; it shows clearly a fine scale structure, the most typical and most remarkable characteristic of high speed flow. The picture is taken using the so-called shadowgraph technique which is particularly sensitive to rapid changes in the density of the gas. The sharp straight lines are shock waves, the grain-like structure turbulence and finally, in the region between the shock the structure looking like twisted spaghetti is due to random acoustic waves emitted by the turbulence.

The tendency of fluid flow toward concentration of gradients of velocity, density, temperature, etc., along sheets or lines is its single most apparent and most interesting feature. The trend is intimately connected with the non-linear character of the equations of motion and most obvious in the breaking of water waves on beaches. In any high speed motion there thus exists a race between the steepening tendency due to the non-linear inertia effects and a smoothing tendency due to the various diffusive phenomena like viscosity, heat conductivity, etc. This race is essential for turbulence, water waves and shock waves. In turbulence it is the concentration and diffusion of vorticity, in water waves the concentration and diffusion of the surface slopes, in shock waves the concentration and diffusion of changes in velocity, density, etc. For turbulence the non-linear aspect

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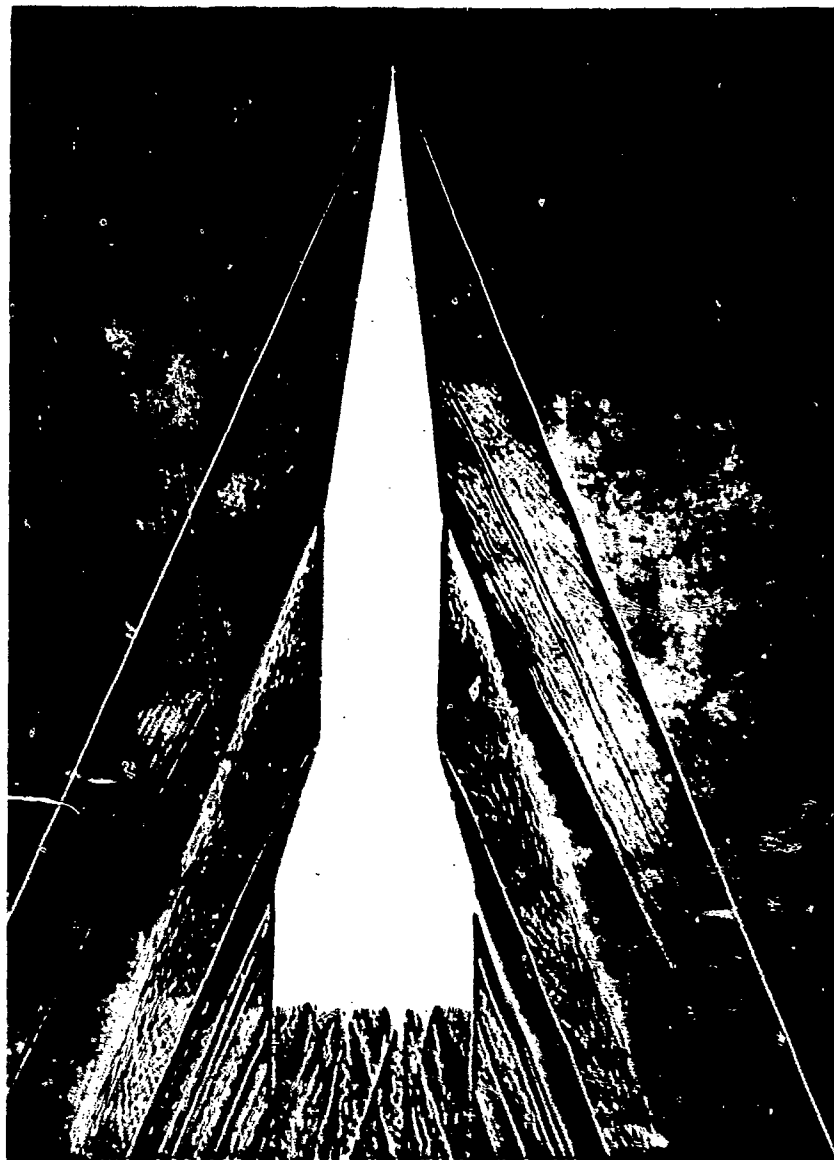
Figure 1. Shock waves, turbulence, and random noise due to a supersonically flying projectile.

of the steepening is more difficult and more interesting, because the concentration of vorticity is an essentially three-dimensional, very involved effect. Shock waves, on the other hand, are basically longitudinal waves, hence can occur and can be studied in one space dimension. The non-linear effect in one dimension can be easily handled, but the diffusive effects which ultimately lead to a definite "shock structure", i.e., a well-defined transition layer, are of primary interest because within a strong shock the gradients can become so large that the usual approach to momentum, heat or matter diffusion becomes inapplicable. The transition layer, i.e., the flow within a shock, thus becomes a test case for attempts to extend hydrodynamic theory into the regions which are far from thermodynamic equilibrium. Even the very simplest thermodynamic working fluid, the perfect, monatomic gas, presents problems and is indeed the one we will discuss here.

PRODUCTION AND PROPAGATION OF SHOCKS

The typical way to make a shock is illustrated in the "piston problem": A piston slides frictionless in an unlimited tube and at time $t = 0$ starts to move, compressing the gas to the right. If the ultimate piston velocity is uniform, say u, then after a while, *regardless of the way u is reached*, a shock wave of unique character propagates ahead of the piston into the undisturbed gas. This "uniqueness" or the permanent nature of the shock means that the "wave" is a transition layer propagating with a fixed velocity c which separates two equilibrium states (1) and (2), with well defined characteristics: ahead of the wave the velocity $u = 0$, the density $\rho = \rho_1$, temperature $T = T_1$, etc. After the wave has passed $u = u$, $\rho = \rho_2$, $T = T_2$, etc.

It is an easy task to relate the variables of state by the so-called "jump conditions". For example the density jump is related to u and c by

$$\frac{\rho_2 - \rho_1}{\rho_1} = \frac{u}{c - u}$$

Two more jump conditions can be obtained from momentum and energy conservation. If one finally adds an equation of state which specifies the substance, for example the gas, in the tube one ends up with a defined problem: Knowing the fluid in the tube and

the piston velocity u, the speed c of the resulting shock wave as well as the conditions existing once it has passed are determined. Note that this can be done without knowledge of any of the transport parameters of the fluid by applying simple thermodynamics and mechanics.

STEEPENING AND DIFFUSION OF VELOCITY GRADIENTS

In speaking about a "wave" or a "transition layer" we imply that the changeover from the undisturbed to the disturbed state takes place rapidly, so that the thickness of the transition zone, δ, is small compared to the tube dimensions. In fact, what does determine δ? Dimensional analysis and similarity require, after all, that δ has to be a multiple of some characteristic length, ℓ, of the problem. Since it does not depend on the tube diameter or any length parameter to be made up from the starting characteristics of the piston, it can only depend upon an intrinsic length parameter of the gas itself. A perfect gas is thermodynamically characterized by its molecular mass, m, only; the molecular "size", the collision cross sections can be neglected. Irreversible transport phenomena like viscosity do depend, of course on the molecular "size", thus depending on the collision cross sections or mean free path of molecular travel between collisions, Λ. Hence it is obvious that the shock thickness depends on Λ or on the transport coefficients, like viscosity, μ, related to Λ

Consequently, a shock wave of a given "strength", measured by its propagation velocity c, the pressure ratio p_2/p_1 or in any other suitable way, will possess a structure determined by the gas and will be independent of the way the shock was produced.

Hence the ultimate balance between inertial steepening and diffusive smoothing is independent of the macroscopic setup of the problem. Indeed the time, t_0, it takes for a velocity profile, which at

$t = 0$ possesses a maximum gradient $s = \dfrac{du}{dx}$, to develop a jump, i.e.,

to make $s \to \infty$ is simply inversely proportional to s, or

$$t_0 \sim 1/s = 1 \Big/ \frac{du}{dx}$$

the time t_1 to smooth, by diffusive action alone, a jump into a profile with a width δ is

$$t_1 = \delta^2/\nu$$

where $\nu = \mu/\rho$ is the ratio of viscosity to density, the so-called kinematic viscosity. Noting that

$$S \doteq \frac{\triangle u}{\delta}$$

the two effects balance if (Fig. 2)

$$\delta \sim \frac{\nu}{\triangle u}$$

Shock Formation

Nonlinear, zero viscosity
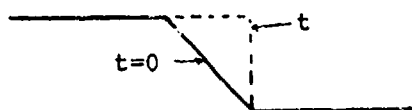
$$t = \frac{2}{\gamma+1} \Big/ \frac{du}{dx} \approx \frac{2}{\gamma+1} \frac{\delta}{\triangle u}$$

linear, finite viscosity

$$\delta = \sqrt{\nu t} \quad \therefore \quad t \approx \frac{\delta^2}{\nu}$$

combined $\qquad \delta \approx \frac{2}{\gamma+1} \frac{\nu}{\triangle u}$

FIGURE 2. The balance of inertia and viscous effects in the shock formation process.

i.e., an estimate of the thickness of the transition zone is easily obtained. For example, for a strong shock propagating into standard air we find

$$\nu_1 \sim .16 \text{ CGS}$$

$$\triangle u = u \sim c$$

Hence if $c = 10a$, a the velocity of sound in air, 3×10^4 cm/sec.

$$\delta \sim \frac{.16}{3 \times 10^5} \sim 5 \times 10^{-7} \text{ cm}$$

This extremely crude result serves mainly to emphasize the dimensions involved. Actually, the computed thickness δ is less than the mean free path Λ and hence our derivation using ν is not self-consistent. Indeed, the reasoning based on ν is adequate, correct in fact, for weak shocks for which $c \to a$ and $\triangle u \to 0$, but for strong shock δ will remain equal to a few mean free paths.

One may add here that one has not found a similar, unique and typical element of turbulence, that is an experimentally realizable "vortex", which demonstrates the non-linear-viscous race in turbulent motion.

ENTROPY

The shock compression and the resulting heating of the gas is a rapid process in which the fluid passes through a state of non-equilibrium within the transition layer. A change of state, in which the material is always close to equilibrium, is reversible in the thermodynamic sense. If during the change of state, non-equilibrium conditions occur in the system, the process is irreversible; the entropy of the system increases. The specific entropy of the gas s, the entropy per unit mass is consequently larger in the gas after the shock has passed, i.e.,

$$s_2 - s_1 > 0$$

and the difference increases with the strength of the shock. Since entropy is a variable of state, $s_2 - s_1$ can be computed without reference to the processes going on in the transition layer. The "production" of entropy occurs, of course, in the transition zone which adjusts its thickness and hence the slopes of the velocity and temperature profiles, to produce the necessary increase in entropy. Within the range of applicability of the classical equation of

hydrodynamics—or for that matter, within the regime of so-called irreversible thermodynamics—the entropy production is proportional to the square of the velocity and temperature gradients,

$$\mu\left(\frac{du}{dx}\right)^2, k\left(\frac{dT}{dx}\right)^2$$ where μ and k are viscosity and heat conductivity

respectively.

Thus the stronger the wave, the thinner the shock, the larger the gradients and the larger the entropy change. However, the thickness change must be such that the total entropy increase becomes independent of the transport parameters μ and k. It is easily seen that this is true if δ scales linearly with μ and k, a result which followed already from the balance of diffusion and steepening.

Beyond the range of applicability of concepts like viscosity and heat conductivity any complete theory must show these general features: An entropy production due to stress and heat flux within the transition zone which depends on molecular interactions involving the range of the intermolecular force fields, i.e., for a gas, the collision cross section. The form of the production terms must be such that the total entropy production becomes independent of the details of the molecular processes.

DISTRIBUTION FUNCTION

We restrict the discussion now to a simple monatomic gas like argon. For such a gas, one can define a distribution function f such that $f(\vec{v}, \vec{x}, t)$ denotes the number of molecules which are found at a given position in space at a given time and with a specified velocity within certain tolerances. f has the character of a probability density or statistical weight, macroscopically measurable quantities are the averages formed using f: For example the number of particles at $\vec{x}$ and t is simply the sum or integral of f summed over all velocities v.

$$n = \int f \, d\vec{v}$$

Similarly the mean velocity $\vec{u}$ at $\vec{x}$ and t is given by

$$nu = \int \vec{v} f \, d\vec{v}$$

The difference between the velocity $\vec{v}$ and the mean velocity $\vec{u}$ is usually called $\vec{C}$

$$\vec{v} - \vec{u} = \vec{C}$$

C is the random component of molecular velocities as seen by an observer traveling along with the mean velocity u.

Knowing f, any macroscopically measurable variable of the gas can be computed. Indeed, f includes much more information than we usually need and it is therefore not too surprising that the determination of f is very difficult. The equation from which f can be obtained in principle is the famous Boltzmann equation: The distribution function is altered by collisions between molecules. Each collision process can be computed in a straightforward—but not always simple—way from mechanics if the initial condition of the two colliding molecules are known.

These initial conditions are considered random parameters; in this way a statistical element was introduced by Boltzmann into the otherwise completely deterministic theory. The equation for the distribution function f can then be written in an innocent looking way

$$\frac{df}{dt} = G - fL$$

stating that the rate of change of f is due to a Gain, G, minus a Loss fL, i.e., molecules in the appropriate velocity range are gained and lost by collisions, f increases or decreases as the net result of these processes. Unfortunately G and L are complicated integrals involving f in a non-linear way.

In equilibrium $\frac{df}{dt} = 0$ and it is easy to show that the distribution function which is now independent of $\vec{x}$ and t reduces to the well-known Maxwellian or Gaussian distribution, $F(\vec{v})$.

If the gas is near equilibrium so that f does not differ much (in some sense!) from F, one can use the so-called Chapman-Enskog theory which shows that the classical hydrodynamical equations are consistent with the first approximations to the Boltzmann

equation, i.e., near equilibrium $f - F$ depends linearly on the gradients of velocity and temperature.

STRESS AND HEAT FLUX

Momentum and energy flow can be written in terms of the molecular velocities C; the momentum carried by each molecule is simply $m\vec{C}$, the volume flow $n\vec{C}$, the momentum flow through unit area in unit time is

$$ nm\overline{\vec{C}\vec{C}} = \vec{\vec{P}} $$

Similarly the flow of energy is equal to the mean product of $\frac{m}{2} C^2$, the energy of a molecule times the volume flow

$$ \frac{nm}{2}\overline{\vec{C}C^2} = \vec{q} $$

In equilibrium, conditions in the gas are uniform, i.e., no direction and no position is distinguished. In this case averages like $\overline{C_1 C_2}$ as well as $\overline{C_1 C_2^2}$ and $\overline{C_1^3}$ are zero. Hence $q = 0$ and P now contains only the three terms

$$ nm\,\overline{C_1^2},\ nm\overline{C_2^2},\ nm\overline{C_3^2} $$

which, moreover, must be equal; the thermodynamic pressure P is thus defined by

$$ P = \frac{1}{3}nm(\overline{C_1^2} + \overline{C_2^2} + \overline{C_3^2}) = \frac{nm}{3}\overline{C^2} $$

Now consider non-equilibrium flow. To make matters simple specialize immediately to a shock layer. Heat flux and viscous stress then have only one component:

$$ q_1 = \frac{nm}{2}\,\overline{C_1 C^2} = \frac{nm}{2}(\overline{C_1^3} + \overline{C_1 C_2^2} + \overline{C_1 C_3^2}) $$

The stress component P_{11} is made up of thermodynamic pressure P and viscous stress τ

$$P_{11} = p + \tau = nm \, \overline{C_1^2}$$

hence

$$\tau = nm(\overline{C_1^2} - \frac{1}{3}\overline{C^2}) = nm \, [\overline{C_1^2} - \frac{1}{3}(\overline{C_1^2} + \overline{C_2^2} + \overline{C_3^2})]$$

By symmetry, the $\overline{C_2^2} = \overline{C_3^2}$ and τ and q can be written

$$\tau = \frac{2}{3} nm(\overline{C_1^2} - \overline{C_2^2})$$

$$q = \frac{nm}{2}(\overline{C_1^3} + 2\overline{C_1 C_2^2})$$

Of particular interest is the ratio of τ/p

$$\tau/p = \frac{2(\overline{C_1^2} - \overline{C_2^2})}{\overline{C_1^2} + 2\overline{C_2^2}}$$

which is a convenient non-dimensional measure for the departure from equilibrium Indeed, τ/p is essentially the expansion parameter in the Chapman-Enskog theory and hence we know that if τ/p is small compared to unity

$$\tau = \mu \frac{du}{dx}$$

$$q = -k \frac{dT}{dx}$$

Hence, as expected for small deviations from equilibrium, we obtain the usual Navier-Stokes equations of fluid dynamics. The transport parameters μ and k are functions of T which, in principle, can be determined for any intermolecular force law.

It is not difficult to show that for strong shocks, $\frac{\tau}{p}$ increases rapidly and exceeds unity, at least in parts of the shock layer. Hence the Chapman-Enskog series cannot be expected to converge. For strong shocks one must therefore find $f(\vec{x}, \vec{v})$ and determine from it the measurable variables of state. Solving the Boltzmann equation exactly is out of question; hence the attempts to find f must be directed toward approximations of a different type than the Chapman-Enskog approach.

For the shock layer problem, two types of approximations can be used: The Mott-Smith method which approximates the distribution function by a weighted sum of the Maxwellians before and after the shock:

$$f = aF_1 + (1 - a)F_2$$

with $a(x)$ a weighting function. $a(x)$ can be determined in such a way that f is an *approximate* solution to the *exact* equation. The other method consists of finding *exact* solutions to *model* equations, i.e., equations which are simpler than the Boltzmann equation but, hopefully, include the essential features. The best known of these model equations is the so-called BGK or Krook model

$$\frac{df}{dt} = An(F - f)$$

in which F is the local Maxwellian. F depends therefore on u, n and T which in turn are moments of the unknown f. The equation is consequently still non-linear but very much simpler than the Boltzmann equation. There is little doubt that the Boltzmann equation contains much more information than is required for a description of an experimentally realizable situation. It is therefore quite reasonable to study simpler model equations like the one by Bhatnagar-Gross-Krook used here.

The model agrees closely with the Navier-Stokes equations if τ/p is small and is compatible with free molecular, i.e., collision-free flow in the opposite limit. It, furthermore, demonstrates at least some of the interesting mathematical properties of the Boltzmann equation in manageable form; in particular the non-uniform convergence of the Chapman-Enskog series is obvious. For the shock wave problem, the model equations can be numerically integrated and all the flow parameters determined. Typical results showing the development of the distribution function through a shock layer are shown in Fig. 3.

EXPERIMENTS

The measurement of the density distribution within a shock layer requires considerable experimental skill and for strong shocks has been successfully carried through only in recent years. The most
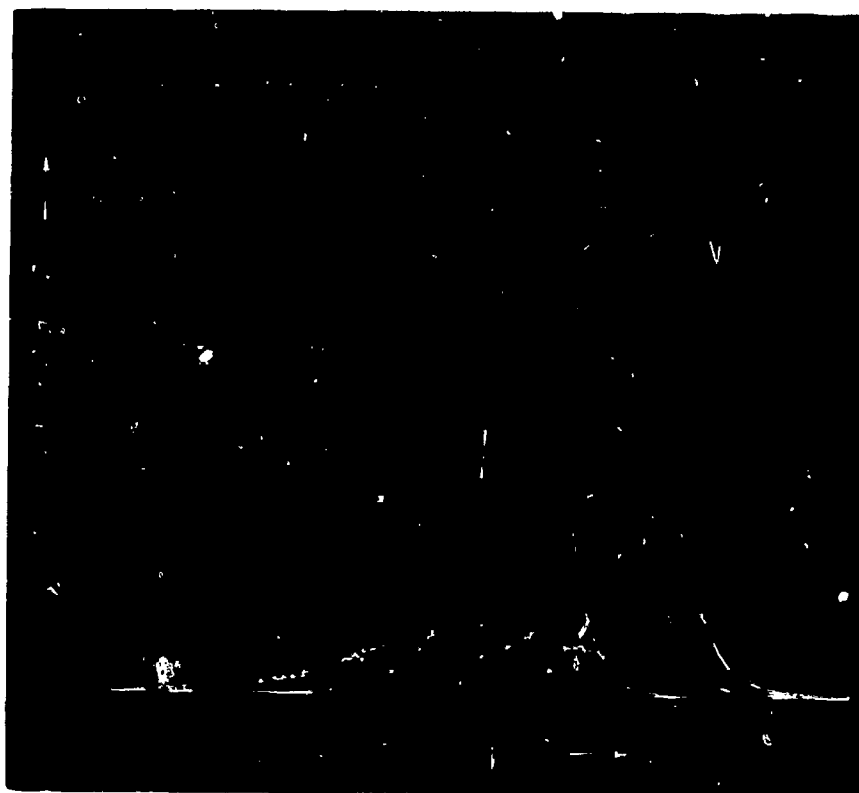
FIGURE 3. Velocity distribution function at various points within a shock wave.

accurate and complete data have been taken with the help of the absorption of an electron beam recently by Schmidt at GALCIT: the principle of the method and sample results are shown in Figs. 4, 5 and 6. The density distribution is compared with BGK computation as well as with the corresponding solution of the Navier-Stokes equations. The latter becomes clearly less and less adequate as the shock strength increases; the overall behavior, e.g., the "thickness" for strong shocks is well represented by the BGK computations. Details in the distribution are, however, different. These differences are significant in pinpointing the various physical phenomena which combine to produce the shock profile. For example, the long upstream precursor of the BGK distribution is qualitatively certainly real and represents the diffusion of "hot", that is, fast

Figure 4. Electron-beam densitometer applied to shock structure measurements in a shock tube.

molecules. It is quantitatively exaggerated because of the molecular model which corresponds to a very "soft" potential.

The use of sophisticated shock tubes and the study of model equations in these recent studies of shock structure have gone a long
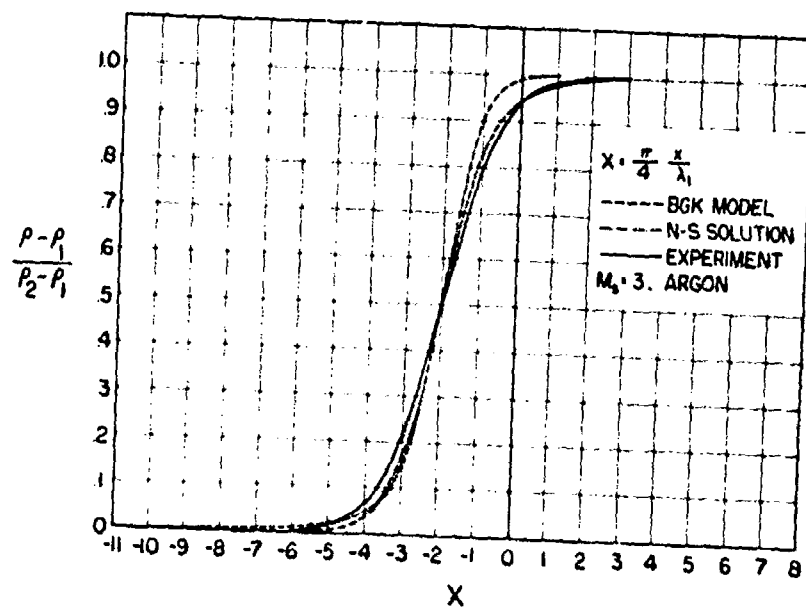
FIGURE 5. Density distribution within a shock wave at $M = 3$, measured and computed.
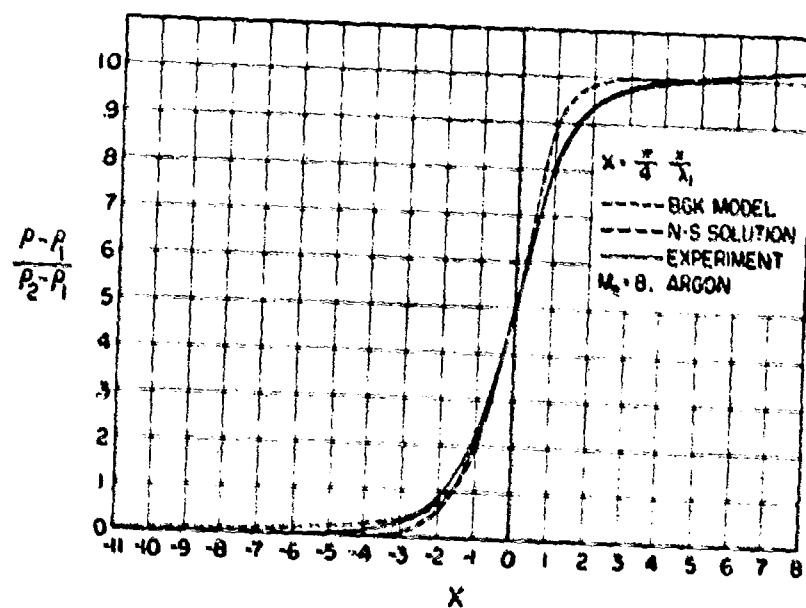
FIGURE 6. Density distribution within a shock wave at $M = 8$, measured and computed.

way in clarifying rarefied gas flows, in particular in the transition zone where neither the Navier-Stokes nor the collisionless approximation holds. In particular, it has become clear why and how the difficulties with the Chapman-Enskog procedures appear if the method is applied far from equilibrium. The interplay of *local* molecular interactions which typically lead to transport proportional to the gradients of the observables and of *global* interactions of fast molecules with long free paths can be studied very successfully using the shock structure as a mathematical and experimental model. It is in this sense that the use of model equations has advantages over, say, the Mott-Smith approximation which can give the shock thickness and the shock profile as well as, or better and with less labor.

IX. Mappings as a Basic Mathematical Concept

SAUNDERS MAC LANE

Mathematical research is sometimes devoted to attacking and solving explicit hard problems; such problems may arise within mathematics itself or in one of its many applications. At other times, mathematical research is concerned with the disclosure and development of new general concepts, especially those which are "abstracted" from particular mathematical situations. In some cases, these abstractions will themselves later lead to explicit hard problems; in all cases, they should lead to a clarification and better understanding, because such ideas, though abstract, can be simple.

This talk will deal with an example of this second type of general development. It is thus a report on a sample of one sort of basic research. The particular example to be discussed is one which we hope will be useful in organizing and codifying mathematical knowledge. This is a task which is pressing today, in view of the rapid rate at which knowledge, mathematical and otherwise, is growing.

The abstract idea to be examined is that of a "diagram of mappings". This might also be called a "block diagram" or "arrow diagram". Samples of such diagrams are:
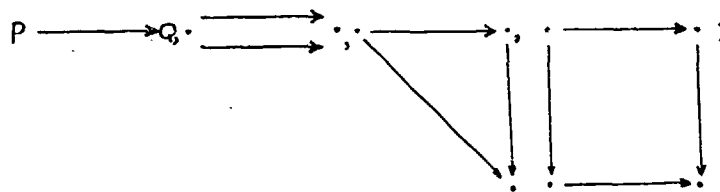
SAUNDERS MAC LANE is the Max Mason Distinguished Service Professor at the University of Chicago where he has served on the faculty in the Mathematics Department since 1947. He is a leading authority on Categorical Algebra, the youngest of the full-fledged fields of mathematics. He has also done extensive research on cohomology theory, topology of function spaces, spectral theory and projective spaces.

There are many others, including much bigger ones (even with an infinite number of different arrows). We call such an arrow diagram a *category* and explain this notion as follows. A category consists of *vertices* P, Q, R, ... and *arrows* f, g ... Each arrow *starts* from a vertex P and *ends* at some vertex Q (which *may* happen to be the same vertex, Q = P). We write f: P → Q to indicate that f starts at P (or, has "domain" P) and ends at Q (or, has "codomain" Q). Most important, the compound path formed by following two successive arrows f and g is always represented by another single arrow c, called the *composite* c of g with f, and written as c = g°f, as in the diagram
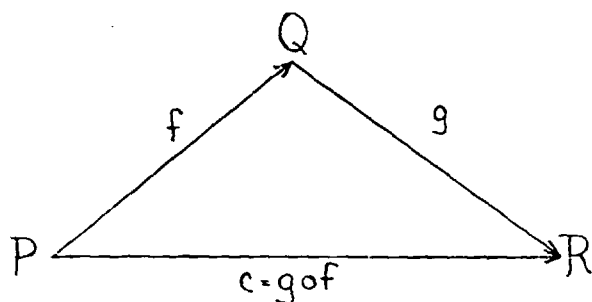
This requirement can be stated more formally as follows: if the arrow g starts where the arrow f ends, then the composite arrow c = g°f is defined; it starts where f does and ends where g does.

Given three arrows f, g, and h in succession, we can then form a triple composite, as in the following figure.
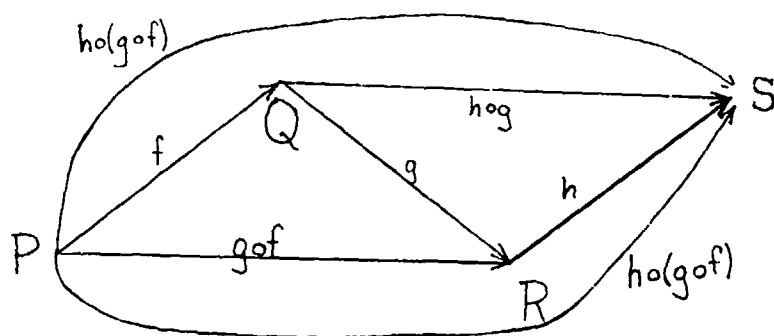
This successive composition can be done in two different ways:

first compose g with f, and then h with the result, or first h with g, and then the result with f, as indicated. We require as an axiom, that the results of these two different triple composites be equal or, in symbols:

$$(1) \qquad\qquad h^\circ\ (g^\circ f) = (h^\circ g)^\circ\ f.$$

This axiom is called the *associative law* for composition, because it is like the associative law $x(yz) = (xy)z$ for the multiplication of numbers x, y, and z.

Another law for the multiplication of numbers is the property of the number 1, which states that $1y = y1$ for any number y; we say that the number 1 is an "identity" for multiplication. Similarly, in an arrow diagram, we assume that for each vertex P there is an *identity arrow* 1_P which starts and ends at P. In the diagram
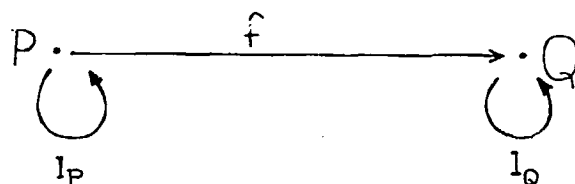
we have indicated two such identity arrows, those at the vertices P and Q. This diagram also suggests the composites which might be formed with such identities; our second axiom now requires that these composites satisfy

$$(2) \qquad\qquad 1_Q\ ^\circ\ f = f = f\ ^\circ\ 1_P.$$

for any arrow f: $P \to Q$. With the axioms (1) and (2) the idea of a *category* (arrow diagram) is completely defined. Now for a few of the many examples.

First, a function may be regarded as an arrow. The usual graph $y = f(x)$ of a function may be considered as a rule which gives to each point x_1 on an x-axis, a corresponding point $y_1 = f(x_1)$ on the y-axis:
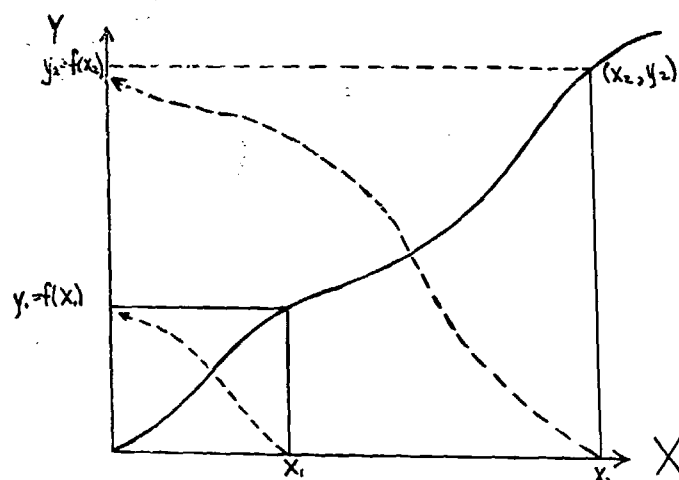
Thus f gives arrows $x_1 \to f(x_1)$, $x_2 \to f(x_2)$, ..., and so "maps" the set X of all points x to the set Y. In general, if X is any set (i.e., any collection of elements) and Y another set, a function f with domain X and range Y (a function on X to Y) is any rule which assigns to each element x of X an element $f(x)$ of Y. We may think of the function f as an arrow f which starts at X and ends at Y. (An arrow built up from all the individual arrows starting at an element x_i of X and ending at the element $f(x_i)$ in Y.) If g is some other function starting at Y and ending, say, at Z, there is an evident way of forming a composite function $c = g \circ f$:
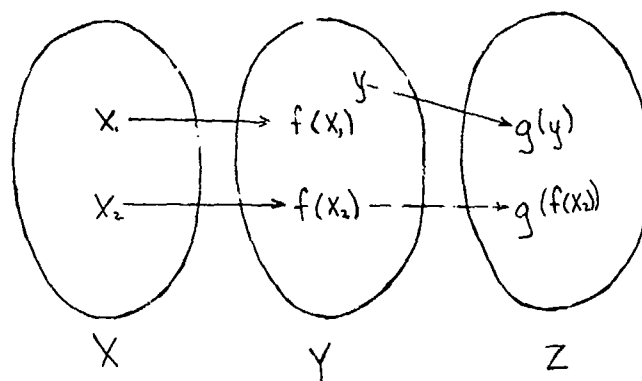
It is the function c which assigns to each element x of X the element $g(f(x))$ of Z— in other words, to apply c, first apply f to x, and then g to the result. This $c = g \circ f$ is the usual composite function.

This defines the *category of sets*. The vertices X, Y, Z of the category are sets, and the arrows f: X → Y of the category are the functions with domain the set X and range Y, while composition of functions is the operation just now defined. Moreover, each set X has an "identity function" 1_x: X → X, which is just the function which sends every element x of X to itself. With this description one may see that the axioms (1) and (2) for a category both hold.

This category of sets may be taken as the starting point for the foundations of mathematics. Recently, the introduction of the "new mathematics" in the schools has emphasized set theory and the possibility of building up all mathematical discourse in terms of sets (especially, using sets to describe finite and infinite cardinal and ordinal numbers). Some experts now hold that the usual description of set theory via sets and their elements is too limited, and might be replaced by suitable axioms on the category of sets (see reference 1 below). This categorical approach ("set theory is obsolete") emphasizes the importance of functions and uses this emphasis to bring out more clearly the structure of mathematics.

Finite categories may also be constructed. The simplest one is the category with just one vertex (say a vertex P) and just one arrow, the identity of this vertex. Another one is the category with two vertices P and Q and three arrows; namely, the two identity arrows plus one arrow a, from P to Q, as in the figure
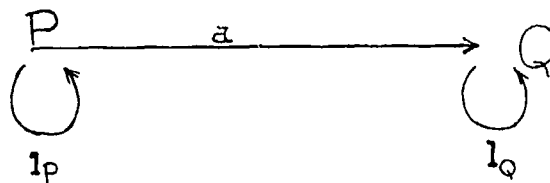
In this category the composites must be defined as $a \circ 1_P = a$, $1_Q \circ a = a$, $1_P \circ 1_P = 1_P$ and $1_Q \circ 1_Q = 1_Q$, all as required by axiom (2). Each of the other sample arrow diagrams listed at the start of this article may be regarded as a category in much the same way (just add an identity arrow at each vertex).

Categories also arise in geometry. For example, the usual rigid motions can be regarded as the arrows of a suitable category. The vertices of this category will be the metric spaces; i.e., the geometrical

spaces where one knows the distance between any two points. In more detail, a *metric space* M is a set of points plus a rule which assigns to each pair of points x, y of M a non-negative real number d(x,y) called the *distance* from x to y. This rule must satisfy two axioms, which require for all points x, y, z that

$$d(x,y) = d(y,x), \quad d(x,y) + d(y,z) \geqq d(x,z).$$

The first states that distance is symmetric, while the second is the triangle axiom:
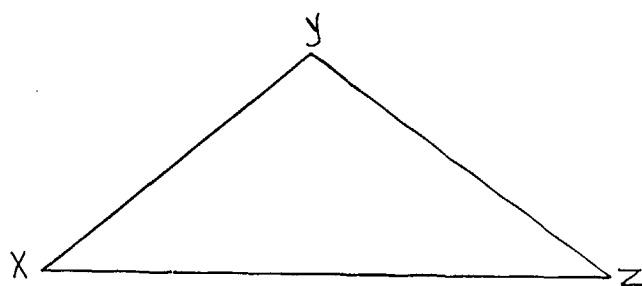
(The "straight" distance d(x,z) from x to z is never longer than that via y). The line, the plane, and 3-space, each with the usual formulas for distances (i.e., for lengths of straight line segments) are metric spaces, as are the Riemannian manifolds of differential geometry, with distance determined from the usual tensor $g_{\lambda\mu}$.

If M and N are metric spaces, a *rigid motion* f from M to N is by definition a function f: M → N which preserves distance, in the sense that

$$d(f(x), f(y)) = d(x,y)$$

for all x and y. This equation states that the distance from x to y is always the same as the distance between the image points f(x) and g(y). (Think of M = N as the plane; then f is one of the usual rigid motions of the plane to itself say, a translation followed by a rotation or by a reflection). In any event, the identity function 1_M: M → M on any metric space M to itself is always a rigid motion in this sense. If two functions f: M → N and g: N → L between metric spaces are both rigid motions, then the function g° f: M → L which is their composite is clearly also a rigid motion. Since the

axioms (1) and (2) hold for the composite so defined, we have described a category, the *category of metric spaces,* with vertices the metric spaces and arrows the rigid motions. Many of the basic properties of metric spaces can be effectively described in terms of this category—for example, the "completion" of a given metric space is a special arrow from the given space to a complete metric space.

This example is typical of the construction of many categories in geometry. Suppose we are studying geometrical objects G, G', G'' of some type, and that an object G of this type is described as a set of points with certain properties and relations. The corresponding category, then, has these objects as its vertices, while an arrow G → G' is just a function from the set G to the set G' which preserves all the listed properties and relations of points of G. Any composite G → G' → G'' of two such functions is another such, as is the identity function 1_G: G → G for any geometrical object of the type considered. Since axioms (1) and (2) hold, we have constructed a category; namely, the category of all geometric objects of the given type. It turns out that many basic geometric constructions may be formulated in perspicuous ways in these categories.

Much the same applies to algebraic objects: one may form appropriate categories in which the vertices are algebraic objects (sets equipped with certain operations) and the arrows are the functions which preserve these operations. As a sample, we consider "monoids" (which are often called "semigroups with identity element"). We define: a *monoid* M is a category with just one vertex P. This means that all the arrows m, n of this category M must start *and* end at P. Hence any two arrows of M have a composite m° n which is another arrow. If we write the composite m° n just as a product mn, then the axioms (1) and (2) for a category simply state that the product is associative and that there is a special element (the arrow 1) which is an identity for this product. In other words, we could have defined a monoid M to be a set (of arrows m, n) with a product mn which is associative and which has an identity element 1. It will be easier to call the m's in M not arrows but *elements* of the monoid M.

The definition of a monoid is much like the definition of a group; a *group* is a monoid in which every equation mx = 1 has a solution x (the solution x = m^{-1} is the "inverse" of the element m).

Now we define the arrows between two monoids M and K. An

arrow f: $M \to K$ will be a function f on the set M to the set K which satisfies

$$(3) \qquad\qquad f(mn) = (f(m))\,(f(n))$$

for all elements m, n of M. This property (3) states that f takes products to products (the product mn to the product (fm) (fn)). One often says instead that f "preserves products" or that f is a "homomorphism" of multiplication. If two functions f: $M \to K$ and g: $K \to L$ between monoids both preserve products, so does their composite $g°f$. For any monoid M, the identity function 1_M: $M \to M$ evidently preserves products. Hence we have constructed a category, the category of monoids, which has all monoids as vertices and all product preserving functions as arrows. There is strong indication that this category of monoids can be used in the theory of automata, since the semigroups have already been used there.

The category of groups is similarly described; the vertices are groups and the arrows f: $G \to H$ are the homomorphisms of groups (that is, the product preserving functions on one group G to another group H). Similar constructions apply to other algebraic systems, taking appropriate account of the operations used to define these systems. Thus for systems described by two operations of addition and multiplication, the arrows (the homomorphisms) are the functions preserving both sums and products. In this fashion one obtains both the category of all rings and that of all fields. Another important category is that of vector spaces: the vertices of the category are the vector spaces V (with real scalars); the arrows f: $V \to W$ are the functions which preserve all sums *and* preserve multiplication by any real scalar (such a function is just a linear transformation; that is, a function given in terms of bases by a matrix of real scalars).

This is closely related to the category of matrices. One can form the product BA of two rectangular matrices of real numbers *only* when the matrices B and A fit: say B is $n \times m$ and A is an $m \times k$ matrix, with the same m. This fact means that the matrices can be viewed as the arrows of a category. The *vertices* of this category are the integers $k = 1,2,3,\ldots$; an $m \times k$ matrix A is an arrow from k to m. The composite of two arrows B: $m \to n$ and A: $k \to m$ can be formed exactly when there is a matrix product BA, and the composite is defined to be exactly this matrix product. Then axiom

(1) for a category is the usual associative law for matrix multiplication. Moreover, there are many identity matrices—one $n \times n$ identity for each size n—and these identities satisfy axiom (2).

Thus in the category of monoids (or of groups, or of rings) the arrows are those functions, called "homomorphisms", which preserve all the algebraic operations involved. Often the arrows of any category are called its "morphisms". Thus for mathematical systems of any sort, the categorical approach always asks: "For this type of system, what are the morphisms?". This has proved especially useful for complex types of systems (e.g., for manifolds with additional auxiliary structures).

A category C itself is a type of Mathematical System. Hence we ask, what is a morphism F: C → D of categories? It ought to be a function from the "elements" of C to those of D, preserving the algebraic operations involved. Now the elements are vertices and arrows, and the operations are composition and the construction of identity arrows. Hence the morphism F must assign to each vertex P of C a vertex F(P) of D and to each arrow f: P → Q of C an arrow F(f): F(P) → F(Q) of D. These assignments must preserve composition and identities; that is, they must satisfy

$$(4) \qquad F(g \circ f) = (Fg) \circ (Ff), \qquad F(1_P) = 1_{FP}$$

whenever the composite $g \circ f$ of the arrows g and f is defined. Such a morphism F of categories is also called a *functor* on C to D. The defining conditions (4) for a functor may be phrased: a functor takes all diagrams in the category C into diagrams of the same shape in D.

A functor is thus a structure-preserving map from one category to another one. The two categories involved may be of very different types; in particular, there is often occasion to consider functors from a category of geometric objects (say, metric spaces, topological spaces, or Riemannian manifolds) to a category of algebraic objects (say vector spaces or groups). Much of the recent development of algebraic topology has been concerned with such functors; for a given space, the fundamental group, the other homotopy groups, the homology groups, and the cohomology groups all provide such functors. Ten years ago this list contained essentially all the known such functors passing from geometry to algebra. In the last ten years the presence of the functorial approach has been a great stimulus

to the study of new functors of this sort. Many new ones have turned up—the so-called generalized homology theories, such as K-theory and "cobordism".

The functorial approach allows a precise description of the way in which a geometric problem can be first formulated and then solved by an algebraic translation of the problem. Steenrod[2] has clearly explained how this occurs with the use of cohomology operations to solve homotopy classification problems. A typical such problem is this: given two (metric or topological) spaces X and Y and a continuous function f: $X \to Y$, when is this function "trivial", in the sense that it can be continuously deformed to another function mapping all of X into a single point in Y? The mechanism of algebraic translation is this: take a functor F on the category of topological spaces to (say) the category of groups. Then F assigns to the given arrow f: $X \to Y$ between spaces an image arrow F(f): $F(X) \to F(Y)$ between groups. It usually follows easily that if f is trivial, F(f) must also be zero (but not vice versa). Hence if we find a functor F with $F(f) \neq 0$, we know that the given arrow f is not trivial. If we find only $F(f) = 0$, we do not know whether or not the original f was trivial; we must search for new functors F with greater powers of discrimination. The development of algebraic topology has indeed led to the successive discovery of many such functors, often found by taking them with values in categories of algebraic objects with successively more elaborate structure.

These examples suggest that the notion of a functor is useful in describing other cases of the translation of problems and their solutions from one context to another. There are, in fact, many such examples. An important one in differential geometry is the functor mapping each "Lie Group" to its "Lie Algebra".

The notion of a "universal mapping" is another general idea closely related to that of a functor. Suppose we are working within some category, and that some of the vertices in this category have an especially desirable good property: let us call these special vertices the "good" gadgets or just the "gadgets" G. Given any vertex P, we want an arrow from P to a gadget. Such an arrow u: $P \to G$ is called a *universal mapping* (for the given class of "good" gadgets) when the following holds: If w: $P \to G'$ is any *other* arrow starting at P and ending at a gadget G', then there is exactly *one* arrow f: $G \to G'$ with $f \circ u = w$. The diagram is

$$, \quad f \circ u = w$$
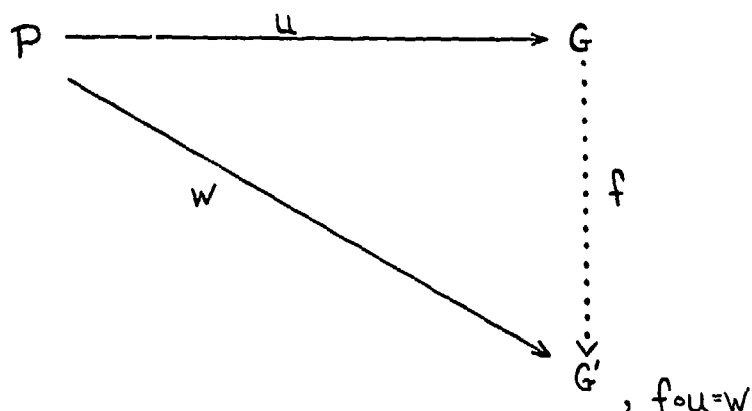
u is universal if to each w there exists (dotted arrow) a unique such f. Put differently: every w factors uniquely, as $w = f \circ u$, through the universal arrow u.

There are many examples of such universal arrows. Some occur in the construction of our number systems. Let us consider a number system to be a set (of numbers) with the two usual algebraic operations of addition and multiplication. An arrow between two such systems will accordingly be a function which preserves both sum and product (i.e., a morphism both of addition and of multiplication). Here are some familiar such systems:

N, all natural numbers	$0, 1, 2, 3, \ldots$
Z, all integers	$0, \pm 1, \pm 2, \pm 3, \ldots$
Q, all rational numbers	n/m for m and $n \neq 0$ integers
R, all real numbers	
C, all complex numbers	$x + iy$, for x and y real.

In this list each system is included in the next, and in each case the inclusion $N \to Z$, $Z \to Q$, etc. can be regarded as an arrow; more explicitly, an arrow in our category of systems with addition and multiplication.

Now in **N**, the system of natural numbers, subtraction is not always possible. (For example, the equation $a + x = 1$ has no solution x.) So let "gadget" mean a system in which subtraction is always possible. Since subtraction is possible in the system **Z** of integers, the inclusion u: $N \to Z$ is a mapping of **N** to a gadget. One can readily show that it is the universal such mapping; any other way of embedding the integers in a gadget (i.e., in a system

with subtraction) can be had by going through **Z**. This provides an exact conceptual description of what **Z** is and reformulates the familiar observation that it is necessary to include all of the integers in any system in which we are to have subtraction (i.e., every negative integer can be described as the solution of some subtraction problem).

In the integers **Z** subtraction is possible, but division by non-zero integers is not always possible: for example, $2x = 1$ does not have an integral solution x. Clearly the rational numbers m/n are introduced exactly in order to have solutions to such equations $nx = m$ in integers m and $n \neq 0$. This can be formulated in the "universal" language as follows: let a "gadget" now be a system in which both subtraction and division (by non-zero numbers) is possible. Then the system **Q** of all rational numbers is a gadget, and moreover, the inclusion u: **Z** → **Q** of the integers in **Q** is a universal mapping. One may prove that any other morphism from **Z** to a gadget must factor uniquely through **Q**.

In the same way one can show that the inclusion mapping **Q** → **R** into the real numbers **R** is universal for mappings from **Q** into systems in which every convergent sequence has a limit. Thus **R** has this property but in **R** the equation $x^2 = -1$ has no root. This equation does have a root in the system **C** of complex numbers (in fact, it has two roots $x = +i$ and $x = -i$ there). This accounts exactly for the complex numbers. In other words, the inclusion **R** → **C** is universal among mappings from the reals to a system where $x^2 = -1$ has a solution.

Usually the complex numbers are defined as numbers $x + iy$, for $i = \sqrt{-1}$ and x and y real numbers, with certain formulas given to define addition and multiplication of these numbers. Alternatively, the complex number $x + iy$ may be described simply as an ordered pair (x,y) composed of two real numbers, again with suitable formulas for sums and products of pairs. This description may be made more geometric by saying that the complex number $x + iy$ is the point with coordinates (x,y) in the plane. As is well known, the operations of addition and multiplication can then be given geometrically. However, all of these descriptions really amount to the same thing, and in every case the rules for addition and multiplication can be deduced from the one fact that the arrow **R** → **C** provides a *universal* solution for $x^2 = -1$. Now, it can

be readily seen that an arrow solving such a universal problem is essentially uniquely determined. Hence the universality of $R \to C$ is a complete description of the complex numbers C in terms of the real numbers.

Stated differently: all the properties of the complex numbers are deduced in this way from the real numbers and the one equation $x^2 = -1$. A corresponding process can be used to construct other number systems. Thus if we start from the system Q of rational numbers and any polynomial equation which cannot be factored there (which is "irreducible" over Q), we obtain a universal construction which gives one of the so-called algebraic number fields. The study of these fields is a flourishing branch of algebra. Again, if we start with any system with appropriate operations of addition and multiplication (technically, one of the systems called a *ring*) and with any suitable collection I of elements of R, we obtain a universal mapping $R \to R/I$ which ends at a new ring R/I, called a quotient ring of R. This mapping is a homomorphism universal among homomorphisms starting from R and trivial on I. (Technically, with kernel containing the "ideal" I.)

Universal constructions of quotient systems like this apply to other parts of algebra. In group theory, one uses extensively the quotient group G/N of a group G by a normal subgroup N of G. This quotient group can be completely described by a universal property of the projections $G \to G/N$ (the projection is that arrow which sends each element of G to its "coset" modulo N). The formulas $(G/N)/(M/N) \cong G/M$ and other "homomorphism" theorems involving these quotients can be proved directly from the universality. A similar description applies to the equivalence classes of a set modulo reflexive, symmetric, and transitive relation, or to the quotient of a space obtained by "collapsing" a subspace to a point.

The familiar process of substituting constants for variables in a polynomial is also an example of universality. To be explicit, take polynomials in a "variable" or "letter" x with integral coefficients, such as the polynomial

$$p(x) = 4 + 7x - 9x^2 + 5x^3.$$

One may substitute $x = 2$ to get $p(2) = 4 + 7 \cdot 2 - 9 \cdot 4 + 5 \cdot 8 = 6$ or $x = m$ for any integer m to get

$$p(m) = 4 + 7m - 9m^2 + 3m^3.$$

The same applies to any other polynomial $q(x)$ with coefficients in the system **Z**. Now all these polynomials $p, q, \ldots$ themselves form an algebraic system under the evident operations of formal addition and multiplication; this system is usually denoted by **Z** [x], and called the system of polynomials in x over **Z**. It includes the constant polynomials (the integers **Z**) as a subsystem; this inclusion i: **Z**→ **Z** [x] is an arrow. The process of substituting the integer m for x in every polynomial $p(x)$, $q(x)$ is also an arrow **Z** [x] → **Z** which sends every polynomial to an integer and the polynomial x to the chosen value m. The corresponding diagram is
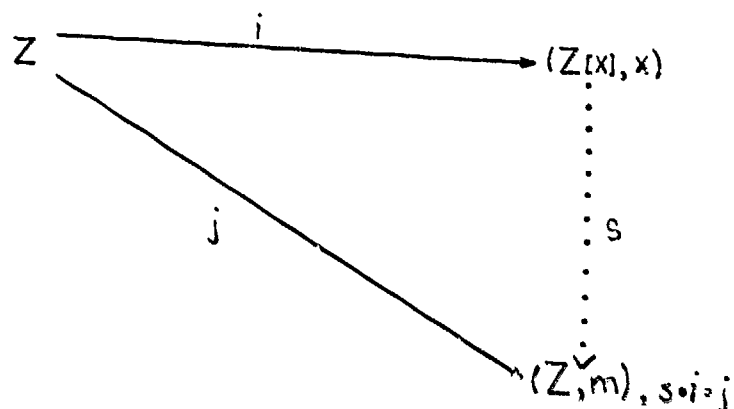
This states that the inclusion i is universal. In detail, let a "gadget" now mean an algebraic system (addition and multiplication) *with* a selected element. Then the system **Z** [x] of polynomials with the polynomial x selected is such a gadget, as is also the system of integers **Z** with m selected. The proposed universal mapping i is an arrow from **Z** to a gadget; any other arrow j from **Z** to the gadget **Z** with m selected has the form $j \equiv s \circ i$, where the arrow s (the process of substitution) carries the selected x to the selected m.

Next consider periodic functions. We shall show that there is a "universal" periodic function, given essentially by taking for each real number x the angle measured by x. (It will be convenient to measure angles in radians, $360° \equiv 2\pi$ radians.) We can formally define: a function $g(x)$ of a real variable x is *periodic*, of period 2π, when $g(x + 2\pi) \equiv g(x)$ for all numbers x. This implies also that $g(x + 4\pi) \equiv g(x)$, $g(x - 2\pi) \equiv g(x)$, and so on. These periodic

functions include all the familiar ones like sin x, tan x, cos 2x, . . .
A periodic function may (like these) take real numbers as values, or
may have values in any other set.

Consider in particular the function p(x) which sends each real
number x to the angle θ with measure x radians, where we exhibit
the angles by the corresponding points on a circle of radius 1
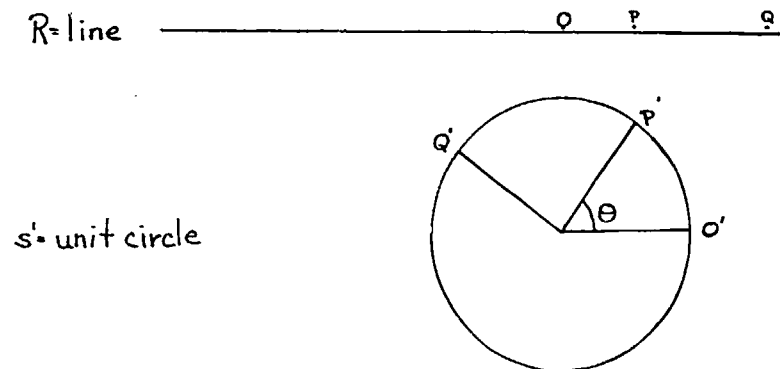
(In the figure, O, P, Q on the real line go to O', P', Q' on the
circle.) Thus p(x) is the function which "wraps" the real line **R**
uniformly around the circle S^1; it is an arrow p: $\mathbf{R} \to S^1$ and is also
clearly periodic: 0. 2π, 4π, . . . are all wrapped by p to the same
point $0' = (1,0)$ on the circle. Now, it is a familiar fact that any
periodic function g(x) can be considered as a function f(θ) of
angles, as in the diagram
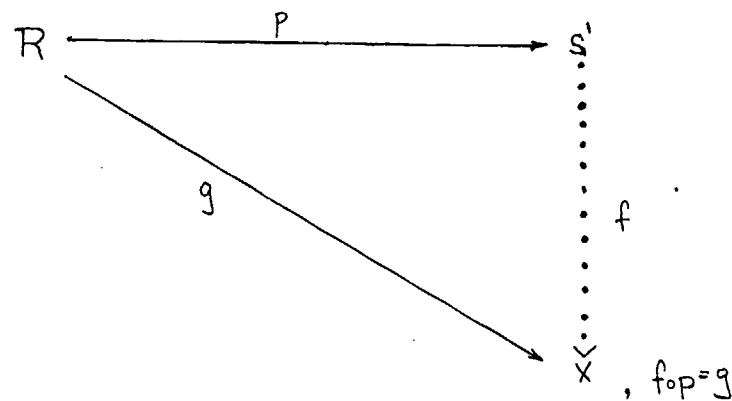
This states that any periodic g can be factored as shown through

the universal periodic function p. In this way familiar facts about angles are examples of universality. Indeed, this universality really can be viewed as a construction of the circle S^1 from the line by the congruence relations "x and x' differ by an integral multiple of 2π."

The basic notion of a subset S of a set X (in symbols $S \subset X$) can be formulated in terms of universality. Take the particular set {0, 1} consisting of just two elements, 0 and 1. Take the subset {0} $\subset$ {0, 1} consisting of 0 alone. This set {0} is a "universal" subset in the following sense. For any subset S of any X we can describe a corresponding "characteristic function" $C_S : X \to \{0,1\}$ with values in the special set {0,1} as the function with

$$C_S(x) = 0 \qquad \text{when x is in S}$$
$$= 1 \qquad \text{when x is not in S.}$$

Now for any function $f: X \to \{0,1\}$ one can describe the "inverse image" of the subset {0} to be the collection of exactly those elements s of X for which $f(s) = 0$ (those elements which map to zero under f). For the characteristic function C_S, the inverse image of {0} is exactly the given subset S, and it is the only function $f: X \to \{0,1\}$ with this inverse image for {0}. This can be exhibited in the figure
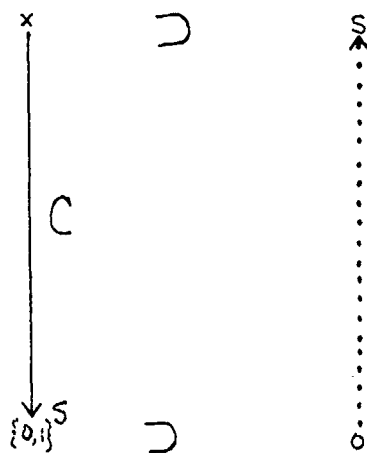
which states (using the "inverse" effect on subsets) that {0} is a universal subset. (The fact that an arrow goes backwards here is typical of "contravariant" effects in many such cases.)

The basic axioms for the arithmetic of natural numbers can also be formulated in terms of a universal mapping. For this purpose, we let a "gadget" consist of a set X together with a function $t: X \to X$ mapping X onto itself. For example, X might be the set $\{0,1,2\}$ with just three elements 0,1,2 while t is the function sending each element to the next (and the last back to 0). Thus the effect of t can be written out as a long sequence

$$0 \to 1 \to 2 \to 0 \to 1 \to 2 \to 0 \ldots \quad .$$

Again, X might be the same set $\{0,1,2\}$ and t the function sending 0 to 1, 1 to 2, and 2 back to 1. The effect of t is then indicated by the following sequence

$$0 \to 1 \to 2 \to 1 \to 2 \to 1 - \ldots \quad .$$

This suggests that the "typical" gadget with a chosen starting point is represented by a sequence (which may or may not repeat itself). The universal such sequence ought to be the sequence

$$\mathbf{N}: 0 \to 1 \to 2 \to 3 \to 4 \to \ldots \quad .$$

This is just the gadget consisting of the set **N** of all natural numbers together with the function $s: \mathbf{N} \to \mathbf{N}$ which sends each natural number n to its "successor" $n + 1$. Now this inclusion of 0 in **N** is an arrow $u: \{0\} \to (\mathbf{N}, s)$ from $\{0\}$ to this gadget **N**. We claim that it is universal: Given any arrow $w: \{0\} \to (X, t)$ from the set $\{0\}$ to a gadget in our present sense, there is a unique function $f: \mathbf{N} \to X$ which is a morphism of these gadgets and which has $f(0) = w(0)$:
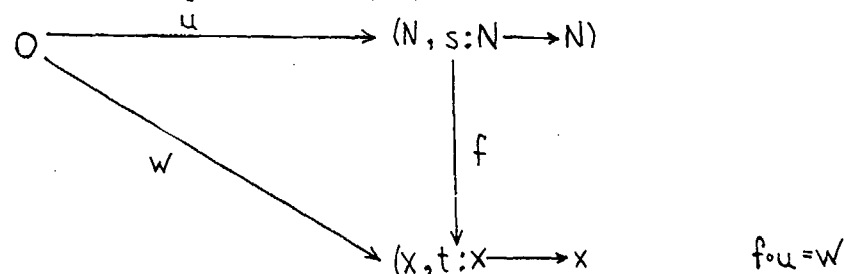
Indeed, to say that $f: \mathbf{N} \to X$ is a map of gadgets means exactly that $f \circ s = t \circ f$, so this map f is given by

$$f(0) = w(0), \ f(1) = t(w0), \ f(2) = t(t(w(0))), \ \ldots$$

and in general $f(m)$ is the result of applying t m times to $w(0)$.

This one property of **N** and the successor function s: $\mathbf{N} \to \mathbf{N}$ can be used as a complete description of the natural numbers. (It is equivalent to the usual Peano postulates for N.) In particular, this property of **N** can be used to construct the operations of addition and subtraction. When combined with other properties, it gives a new system[1] of axioms for set theory.

We have thus given a number of examples of universal mappings: the basic property just given for the system of natural numbers; the arrows including this system in successively larger systems of integers, rational numbers, real numbers, and complex numbers; the arrows mapping a system on a quotient system, or constructing polynomials, or describing subsets by characteristic functions, or mapping numbers upon angles so as to get a universal periodic function. There are many other examples of universal arrows in other parts of mathematics, as well as examples of a related notion called an "adjoint" pair of functors, and there is already a considerable roster of theorems about such functors.

Indeed, functors occur naturally in connection with universal mappings u: $P \to G$. The given class of good gadgets G forms a category C with vertices the gadgets G and arrows the arrows f: $G \to G'$. There is a functor F on this category to the category of sets and described in terms of the given vertex P: This functor assigns to each gadget G the set of all arrows v: $P \to G$ from the fixed P to this G, and to each arrow f: $G \to G'$ the function $F(f):F(G) \to F(G')$ which sends each arrow v to $f \circ v$: $P \to G'$. The universal arrow u: $P \to G$ may be described as a universal element of this functor; this description is effectively used in proving theorems about universal arrows and other adjoint functors. For example, the reader might try to prove that, given P, the universal mapping u: $P \to G$ is essentially unique. Here "essentially" means the following: If u^*: $P \to G^*$ is another universal mapping to some other good gadget G^*, then G^* is isomorphic to G in the exact sense that there are arrows f: $G \to G^*$ and f^*: $G^* \to G$ with $f \circ u = u^*$ and $f^* \circ u^* = u$ as well as

$$f \circ f^* = 1: G^* \to G^*, f^* \circ f = 1: G \to G.$$

These last equations state that f^* is the (two-sided) inverse of f.

With this we end our introduction to mappings and categories.

The subject of category theory and adjoint functors is growing so rapidly that printed information is largely out-of-date, but there are a number of available further sources. An elementary description of the universal elements of functors and their manifold applications to algebra may be found in a text by Mac Lane and Birkhoff.[3] For a more inclusive presentation of category theory one may consult the sprightly monograph by Freyd,[4] the systematic treatment by Mitchell,[5] or the expository article by Mac Lane.[6] The state of present day research on the subject may be sampled in the report on a recent AFOSR-sponsored conference.[7] The applications of category theory to be found there range from topology through algebra to automata. Many more applications and developments are to be expected.

At the coming summer meeting of the American Mathematical Society, the major series of lectures (the Colloquium lectures) will be given by Samuel Eilenberg, on the application of categories and functors to automata. Current investigation of Lawvere (not yet published) indicate a real possibility of a "categorical dynamics" in which many of the standard concepts of dynamics can be formulated in a simpler and more powerful form.

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X. Photochemical Systems

George S. Hammond

I believe that photochemical changes, chemical changes caused by absorption of light, have great potential for use as working parts of complex systems. Chemical reactions in general have great promise for use as gears or driving parts in machines, so photochemistry is only a special case with especially attractive features of fast response and precision control. To me it is amazing that increasing understanding of biochemical processes has not inspired more serious thought about the construction of man-made chemical machines. I do not have in mind just mimicking the chemical machinery of living systems but believe that there is real merit in using the example to illustrate the complexity and compactness available in chemical systems. We already have outstanding examples of the use of chemical change in complex systems; the lead storage battery is a familiar case. However, the general concept has seldom been spelled out and considered seriously.

In this lecture I plan to do two things, both in a rather superficial manner. First, I will describe some of the types of physical and chemical processes that seem important in photochemical systems. In addition, I will add some thoughts concerning conceivable applications in useful systems. I should note that a principal interest in photochemistry to chemists is still development of new and interesting synthetic methods. Applications to synthesis will not be stressed in this presentation, merely because the inviting prospects of synthetic photochemistry are already well-recognized.

In Table I we have listed very briefly some uses and significances

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of photochemistry, in addition to application in chemical synthesis. Monitoring of light is an obvious use which can be put to good service in control systems and can also be used to gather information concerning the intensity and character of the light bath in places that may be somewhat inaccessible: for example, in outer space.

TABLE I.   Significance of Photochemistry
1. Monitoring of light
    a. Intensity
    b. Wavelengths
2. Effects on materials
3. Utilization of solar energy

There are a number of ways in which monitoring can be done. First of all, measurement of total chemical response can provide either an integral or differential measure of light intensity. Systems that undergo irreversible photochemical change will give a permanent record of the total amount of activating light received by a sample. On the other hand, systems that are highly reversible reach stationary states in which a chemical reaction $A \rightarrow B$ is forced by absorption of light and the reverse process $B \rightarrow A$ is an ordinary thermal reaction that attempts to force reversion to A. The ratio A to B at a stationary state therefore can provide a measure of the instantaneous intensity of illumination.

Both kinds of monitoring can be made either selective or non-selective with respect to the wavelength of the exciting light. This, of course, arises from the fact that most photochemically active compounds have distinctive absorption spectra throughout the visible and ultraviolet spectral regions. Obviously, if one wants to gain specific information concerning a particular spectral region one has only to choose a photochemical monitor that absorbs exclusively, or nearly exclusively, in that region. The spectral characteristics of the monitoring system can be supplemented by use of suitable chemical or optical light filters.

The second category listed in Table I is enormously broad, as are most subjects entitled "effects". A good many of the well-known effects of light are the damaging consequences of exposure of many kinds of materials to sunlight. These, of course, include effects on

people; sunburns and skin cancers are two well-known examples of damage to people caused by photochemistry. Effects on other materials are also numerous. Nearly everyone, and certainly most people living in Southern California, are acutely aware of the fact that bright sunlight is responsible for triggering the oxidation reactions that turn organic vapors into the noxious mixture known as smog. In a less dramatic way, photochemical oxidation initiates changes in most common organic materials that we encounter in everyday life; for example, the working lifetimes of many synthetic fibers, plastic films, paints, and varnishes might be extended almost indefinitely if they were always kept in the dark. However, I would not want to leave the impression that "effects" are all undesirable. Amazingly useful effects can be caused by exposure of some kinds of substances to light. An intriguing example is the phenomenon of photoconductivity. Some substances which are normally nonconducting become excellent electrical conductors when exposed to light.

Finally, I will admit to having some fairly grandiose notions concerning the possibility of using man-contrived photochemistry to harvest and store solar energy. I do not have in mind the simple replication of the photosynthetic process of green plants but think in terms of development of new photochemical systems, which will supplement the contribution of natural photosynthesis by being applicable under conditions where the green plant does not fare well. Good examples can be found on much of the surface of this planet, since the total dependence of green plants on water prohibits our getting much useful yield from solar energy that falls on arid portions of the earth's surface. Another even more glamorous example is found in the conditions existing in most places removed from the surface of the earth. For example, if we ever do colonize the moon we will have to establish some sort of a selfcontained energy economy there. As far as I can see, there are only two likely candidates as a basis for that economy. First, and perhaps most likely, is the tapping of nuclear energy, but the only other prospect that I can take seriously is manmade photochemical systems which are not dependent upon the vulgar use of water. Incidentally, such systems will probably be designed to exploit the high-energy light from the sun that does not pass through the earth's atmosphere. Even if green plants could be trained to survive without water, there is considerable doubt as to

whether or not they could be educated to take advantage of the light of wavelengths that bathe the surface of the moon generously but never reach the surface of the earth.

Photochemistry, of course, begins with the absorption of light. In order to discuss the absorption process it is necessary to consider in a little detail what happens to a molecule when it acquires a quantum of light energy. Figure 1 shows very schematically the kind of thinking that we use in analyzing the absorption process and its relationship to ultimate chemical change. First of all, the energy gained by an absorbing system is usually highly localized in individual molecules. So, we can discuss the process of excitation in terms of the excited states of molecules. Most of the excitation energy is acquired by electrons, and the process is therefore referred to as electronic excitation. This is true of visible and ultraviolet light, which are the only sources involved in any known photochemical change. Infrared radiation is absorbed by molecules but goes into exciting vibrations and rotations and is very rapidly lost into the thermal pool of the system. In short, infrared radiation is really only useful for warming up an entire system and does not selectively promote individual molecules to very high levels of excitation.

In Figure 1 the symbol S_0 is used to indicate the ground state of a molecule. The S stands for "singlet". This simply means that most common molecules contain even numbers of electrons and those electrons have their spins paired. The following is a schematic presentation of the usual significance of the terms singlet and triplet, which are commonly encountered in discussions of the mechanisms of photochemical reactions.

Singlets and Triplets
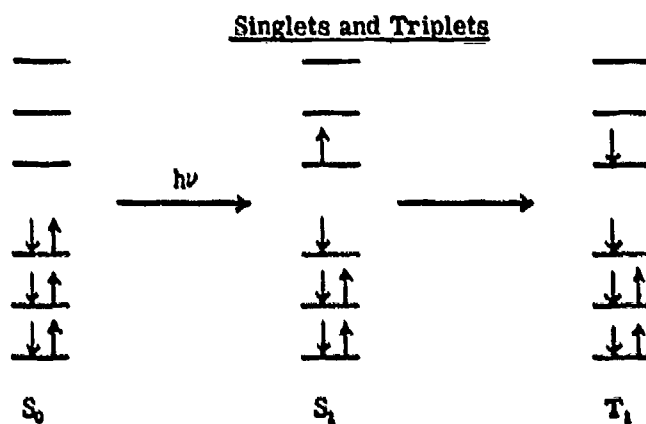
Molecules are described by assigning electrons to orbitals in pairs. The assignment is done so as to use the orbitals of the system having the lowest energy. These orbitals may be of various kinds; such as localized bonding orbitals in a molecule such as methane, or nonbinding orbitals such as those occupied by nonbonding electrons in a water molecule, or they may be delocalized molecular orbitals covering large portions of a molecule as are found in highly unsaturated compounds such as benzene. For our present purposes, it is not
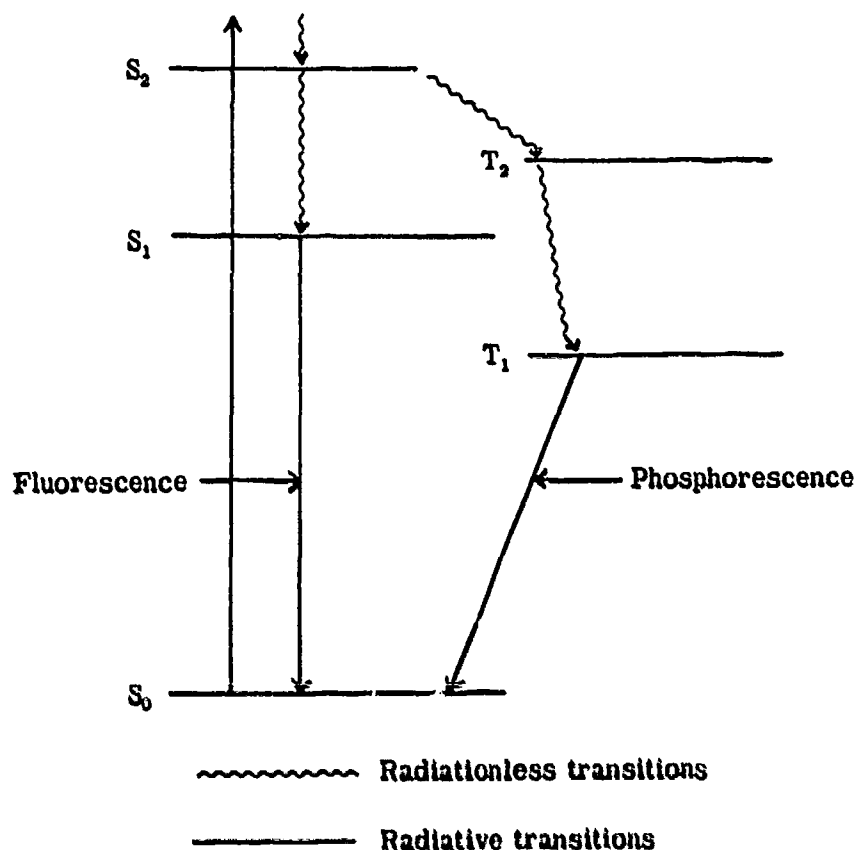
Figure 1. Excitation Processes.

very important to worry about the detailed description of the orbitals involved in excitation. However, development of viable theories concerning the relationship between structure and reactivity in photochemistry will require intimate understanding of such characteristics. As is shown above, absorption of light promotes an electron from an

occupied orbital to one that is unused in the ground state of the molecule. The excited state is designated as S_1, if it is the lowest lying excited singlet state of the molecule. The act of absorption of light by a singlet molecule ordinarily leads only to the production of excited singlet states as primary products. This is one of the very good selection rules that has been known for a very long time by spectroscopists. For all practical purposes, it is completely valid for molecules that contain only light atoms. This includes the majority of organic compounds which are currently of most interest to photochemists. As I have shown above, an excited singlet state has electrons which are orbitally unpaired but still have paired spins. Since the two unpaired electrons occupy different orbitals, they become free to assume parallel as well as antiparallel spin states. If one of the electrons undergoes a spin flip so that the pair spin in the same direction, the molecule as a whole will possess an electronic angular momentum. In a strong magnetic field, the magnetic moment of such a molecule can assume one of three distinctly different orientations with respect to the direction of the applied field. There are really three states rather than one, and this is the reason that the configuration that I have labeled above as T_1 is called a triplet.

Let us return to Figure 1 and trace the fate of a molecule that is excited by absorption of short wavelength light to some excited state having a higher energy content than the lowest excited singlet. Following the course of the straight arrow in the figure, we will produce an S_2 molecule and, along with the electronic excitation, we will put in a certain amount of vibrational energy. In other words, S_2 is not only electronically excited but is also vibrationally excited. This multiply excited species will then undergo very rapid decay processes in which some of the excitation is transferred to other components of the system and simply appears as thermal energy. The fastest of these relaxation rates is probably vibrational energy transfer represented by the short wavy arrow at the top of the figure. This produces a vibrationally unexcited S_2 molecule. Such a state may occasionally live long enough to undergo an *intersystem crossing* to produce an excited triplet, such as T_2. However, the most common fate is simple shedding of electronic excitation in a process called *internal conversion* with the production of an S_1 molecule.

One of the remarkable phenomena of excited state chemistry is

the extraordinary speed with which higher excited states lose some of their excitation and relax to the lowest excited singlet state. Fortunately for photochemistry, the lifetimes of S_1 states are often much longer. These molecules frequently live long enough to emit light called fluorescence or to undergo intersystem crossing to form triplets and, in some cases, take part in chemical reactions. However, the chemically most significant fate of S_1 is intersystem crossing to triplets. The T_1 state of most molecules turns out to be, by a very wide margin, the longest-lived electronically excited state. This is because return from T_1 to S_0 requires not only transfer of a large amount of energy to other parts of the system but also a change in the spin state of one of the electrons. Nature finds the changing of electrical spin functions awkward, irrespective of whether or not the change is radiative or nonradiative. The radiative decay of excited triplet states is sometimes observed and is responsible for the long-lived emission of many molecules called phosphorescence. Study of the phosphorescence spectra provides the most generally valuable tool for gathering information concerning the structure of triplet states.

In addition to the processes indicated in Figure 1, one should add chemical change and transfer of electronic excitation to other molecules. The former is, of course, photochemistry. Electronically excited molecules have very large internal energy contents and are, therefore, potentially capable of undergoing many chemical changes. These may include both unimolecular reactions in which the excited state simply falls apart or turns into some new compound, or the reaction may be bimolecular. In bimolecular reactions the excited state usually reacts with some other molecule in its ground state, although reactions between two excited states are occasionally observed.

The transfer of electronic excitation is most easily detected when the energy acceptor is different from the molecule that originally absorbed the exciting light. Transfer can be readily studied if the energy acceptor undergoes some characteristic reaction or if light emission from the acceptor can be observed. When energy transfer results in chemical transformation of the acceptor, the process is usually called a photosensitized reaction. The following is a generalized mechanism for photosensitized reactions:

$$^1S_0 \xrightarrow{h\nu} {}^1S_n \rightsquigarrow {}^1S_1$$

$$^1S_1 + {}^1A_0 \xrightarrow{k_{st}} {}^1S_0 + {}^1A_1$$

$$^1S_1 \xrightarrow{k_{is}} {}^3S_1$$

$$^3S_1 + {}^1A_0 \xrightarrow{k_{tt}} {}^3A_1 + {}^1S_0$$

$$^1A_1 \text{ or } {}^3A_1 \xrightarrow{k_r} \text{Chemical Reaction}$$
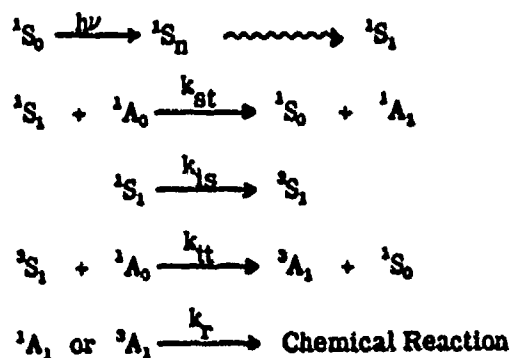
In our own research group, we have been very much interested in the study of sensitized reactions, both because they are inherently intriguing and because they offer very attractive prospects for control and manipulation of photochemistry. For example, we can sometimes make excited states of acceptor molecules, even though they cannot be produced by direct optical excitation of the acceptor. A good example is the case of butadiene:

$$CH_2=CHCH=CH_2$$

If butadiene absorbs light it is promoted to an excited singlet state, but decay directly back to the singlet ground state is so rapid that the molecule does not undergo intersystem crossing to a triplet state with any detectable efficiency. However, excited triplet states of butadiene can be made very readily by energy transfer from the excited triplets of any one of a large number of photosensitizers.

A rather intriguing method for study of energy transfer can be based upon the use of bichromophoric molecules in which there are two nearly isolated, unsaturated units. A few years ago in collaboration with Dr. Peter Leermakers and his group at Wesleyan University, we used both spectroscopic and chemical methods to study internal transfer in a series of compounds containing both a naphthalene-like unit and a unit resembling benzophenone, a common photosensitizer.

Figure 2 shows the ultraviolet absorption spectrum of one of our bichromophoric compounds. The spectrum looks very much like that obtained from a mixture of naphthalene and benzophenone, or more properly 1-methylnaphthalene and 4-methylbenzophenone.
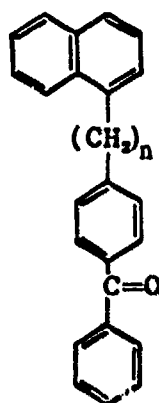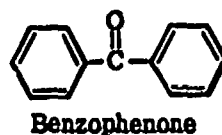
Bichromophoric series, n = 1, 2, 3

The broken line in Figure 2 shows the deviation of the spectrum from that calculated assuming that the two unsaturated units do not interact at all during the process of light absorption. It is obvious that, although there is a small amount of absorption in addition to what might have been predicted, there is no evidence of any very large interaction. However, the emission spectra of the complex molecules are quite different from those of the simple models.

Naphthalene and most of its simple derivatives emit fluorescence after light absorption. Furthermore, after solutions are cooled to sufficiently low temperatures so that they become rigid glasses, naphthalenes also show strong phosphorescence emission arising from the lowest excited triplet state. Benzophenone, on the other hand, shows no detectable fluorescence, but in glassy matrices it has a very strong phosphorescence. Obviously, both compounds undergo intersystem crossing from excited singlets to triplets. Furthermore, it appears that the intersystem crossing of benzophenone is so rapid that it entirely precludes fluorescence.

In Figure 3 we show the phosphorescence emission obtained from methylnaphthalene, from a mixture of methylnaphthalene and methylbenzophenone, and from one of the bichromophoric compounds. Phosphorescence from the two-component mixture is
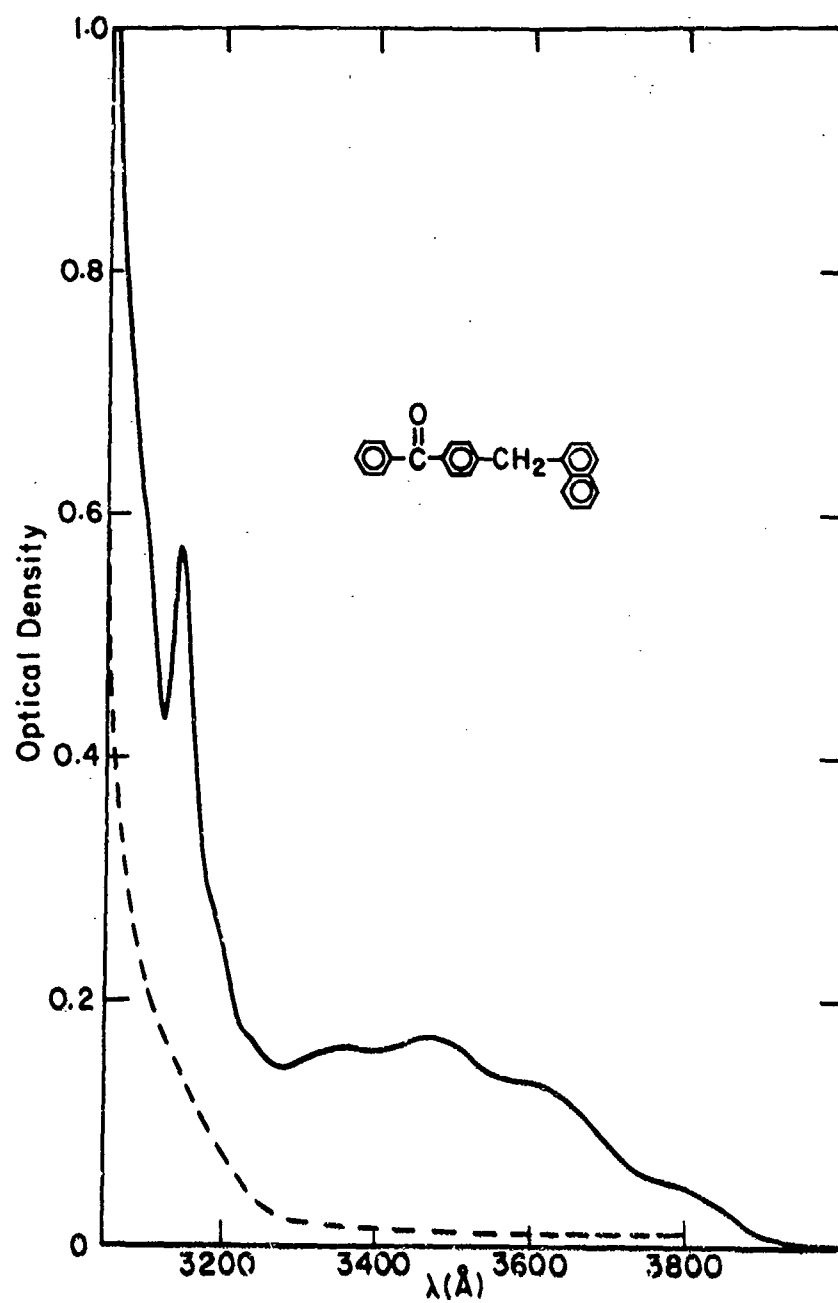
FIGURE 2.
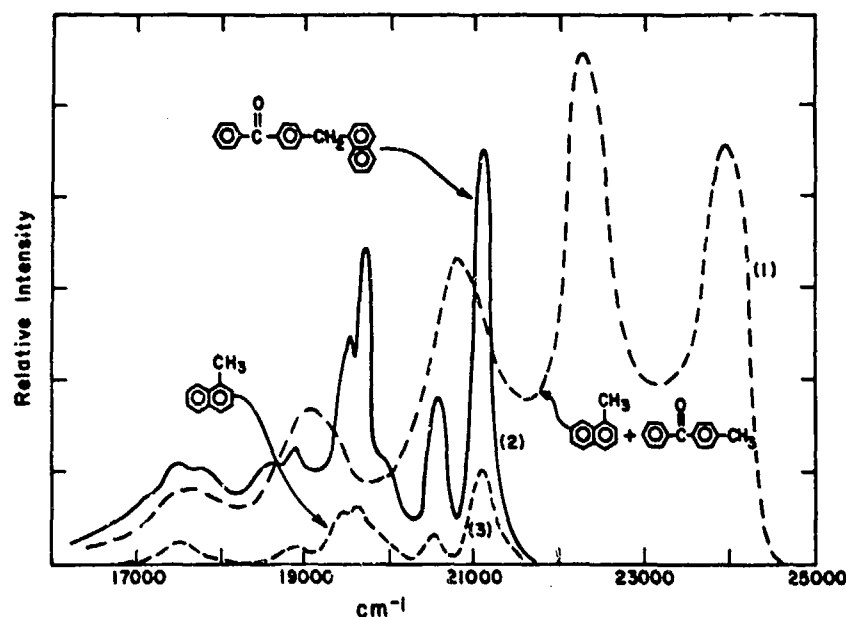
FIGURE 3.

identical to that expected from the two substances acting entirely independently of one another. The large peaks shown in the trace are the characteristic highly structured emission from benzophenone and its simple derivatives. Notice that this emission is completely absent from the spectrum of the bichromophoric material. In fact, the latter has an emission that matches almost exactly the phosphorescence of methylnaphthalene. If we look at a higher frequency where fluorescence of naphthalene is ordinarily found, we find a very weak emission from the complex molecule that looks like highly attenuated naphthalene fluorescence. This is not shown in the figure because the emission is so very small in comparison to either unperturbed naphthalene fluorescence or the strong phosphorescence emission.

The story that we tell concerning the fate of the light energy put into the bichromophoric compound is as follows: light is absorbed by the naphthalene unit and produces an excited singlet state that is essentially naphthalene-like in character. This state has a sufficient lifetime to allow it to emit a very small amount of light. However, most of the energy is rapidly lost by being transferred to the

other chromophore. There it produces an excited state that is essentially the same as the excited singlet state of benzophenone. Intersystem crossing to produce a benzophenone-like triplet occurs very rapidly, just as it would in benzophenone itself. However, the energy is then very quickly transferred back to the naphthalene units forming naphthalene-like triplets. Light is then emitted in a spectrum which matches almost exactly the phosphorescence spectrum of methylnaphthalene. This double shuttling of energy is possible because the separation between naphthalene singlet and triplet states is very much larger than the splitting between benzophenone singlet and triplet states. The following diagram shows the way in which each of the energy transfers can be energetically downhill.

Naphthalene   Benzophenone
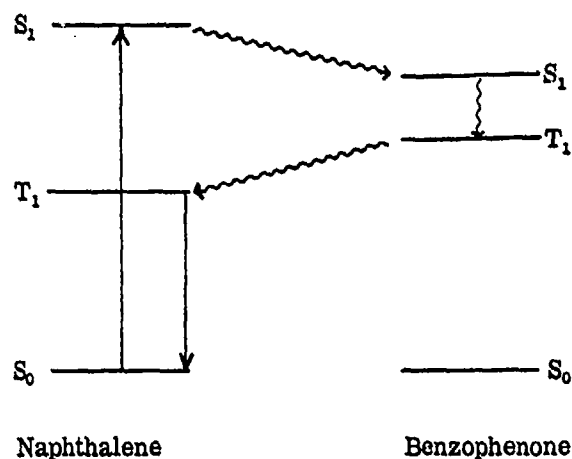
Most of our study of energy transfer has involved bimolecular energy exchange rather than internal shuttling of the excitation. In this work we normally use the quantum yields of photosensitized reactions as a primary tool. However, there is another scheme which is interesting and shows some rather dramatic results. The following equations show a commonly expected mechanism for photosensitized interconversion of *cis* and *trans* isomers of olefinic compounds.

$$S^{*(3)} \;+\; \underset{\underline{\text{trans}}}{\text{R}\diagdown\text{C}{=}\text{C}\diagup\text{H}\;\;\text{H}\diagup\;\;\diagdown\text{R}} \;\;\xrightarrow[\substack{\text{substrate}\\\text{triplet}}]{k_1}\;\; T \;+\; S \qquad (1)$$

$$S^{*(3)} + \underset{\underline{cis}}{\overset{\underset{H}{\overset{R}{\diagdown}}C=C\overset{R}{\underset{H}{\diagup}}}{}} \quad \xrightarrow{k_2} \quad T + S \qquad (2)$$

$$T \xrightarrow{k_3} \underline{trans} \qquad (3)$$

$$T \xrightarrow{k_4} \underline{cis} \qquad (4)$$
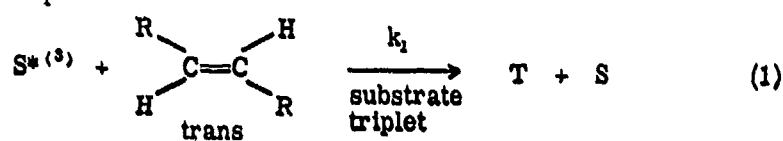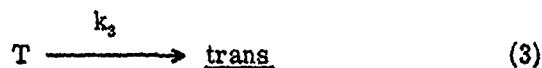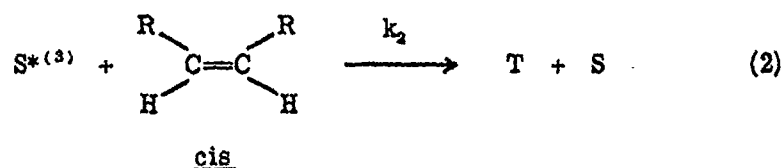
In such a system prolonged irradiation leads to establishment of a "photostationary state". This state, which is sometimes referred to as a photoequilibrium, represents a situation in which the rates of the two opposing reactions have been made equal.

$$R_{c \to t} = R_{t \to c}$$

The composition of the system at the stationary state depends upon the relative reactivities of the *cis* and *trans* isomers as energy acceptors and on the relative amounts of the two compounds formed by decay of the excited triplet state.

$$\frac{[\underline{cis}]_s}{[\underline{trans}]_s} = \boxed{\frac{k_1}{k_2}} \times \boxed{\frac{k_4}{k_3}} \qquad (5)$$

$$\underset{\text{Excitation}}{\underset{\text{Ratio}}{}} \qquad \underset{\text{Decay}}{\underset{\text{Ratio}}{}}$$

The photostationary mixture is obviously a complex function of reactivity factors. What we would like to obtain from its study is information concerning the variation in the rates of energy transfer, k_1 and k_2. We have used a trick which seems to be partially successful in disentangling the complex stationary state relationship. First of all, it is observed in this and many other kinds of studies that the rate of energy transfer is diffusion controlled in solution if the excitation energy available in the donor exceeds by more than a few kilocalories per mole the energy required to promote the acceptor

to its lowest excited triplet state. This is shown schematically in Figure 4. Consequently, for high-energy sensitizers we expect that k_1 will be equal to k_2 and that the excitation ratio will therefore be unity. In such cases, the photostationary state composition should provide a direct measure of the decay ratio $\dfrac{k_4}{k_3}$. This expectation seems to be verified by experiment. The photostationary state composition of a number of substrates is essentially invariant although

$$k_{tt} \cong 5 \times 10^9 \ \ell \ mole^{-1} \ sec^{-1}$$

if $\Delta E_S \geq \Delta E_A + 3\text{-}5 \ kcal \ mole^{-1}$
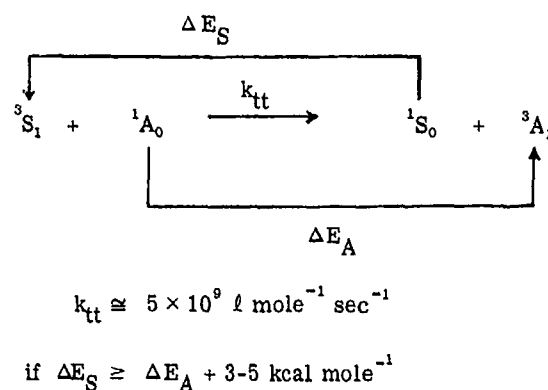
FIGURE 4. Reactivity in Triplet Energy Transfer.

many different high-energy sensitizers have been used to establish it. We can then play the game backwards and assume that the decay ratio remains unchanged when we use sensitizers having lower excitation energies. Changes in the photostationary state will then tell us how the excitation ratio changes as the energy available in the sensitizer is decreased. We can make a straightforward prediction of what will happen, as is shown in Figure 5.

Figure 5 is drawn for a case in which the energy required to excite the *cis* isomer is higher than that needed to excite the *trans* compound. As we lower the sensitizer energy, by varying the structure of the sensitizers we expect that the nature of the transfer to the *cis* isomer will begin to fall off while the rate of transfer to the *trans* isomer is still at, or close to, the diffusion-controlled limit. The photostationary mixtures will then become *cis*-rich and the system will be optically pumped in the *trans→cis* direction. This trend should continue until the excitation energy of the *trans* isomer is ap-

(1) k_{-c}/k_{-t} independent of nature of S

(2) k_t/k_c nearly invariant with high energy S

(3) k_t/k_c variable with low energy S

for $\Delta E_c > \Delta E_t$
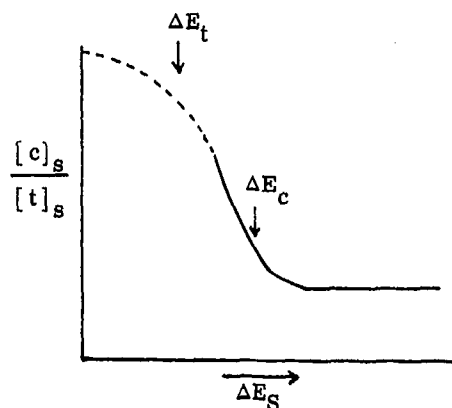
FIGURE 5. Predictions

proached. The efficiency of transfer to the *trans* compound should then begin to fall off. If the energy deficiency has to be supplied as a simple thermal activation energy, the rates of the two transfer reactions should fall off in the same way, after the sensitizers have become deficient with respect to both excitation processes. This should mean that the stationary states observed with low energy sensitizers will again become invariant and probably very rich in the *trans* isomer.

Unfortunately, this naive theory turns out to be woefully incomplete. Figure 6 shows data obtained in the measurement of photostationary states established between the two isomeric stilbenes.

cis-stilbene

$E_T = 57$ kcal/mole

trans-stilbene

$E_T = 49$ kcal/mole
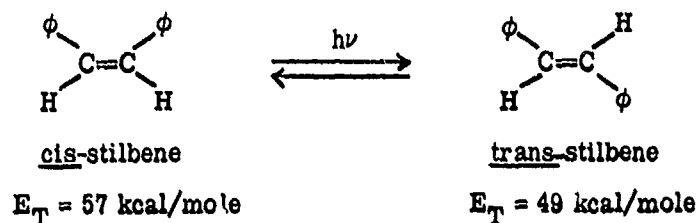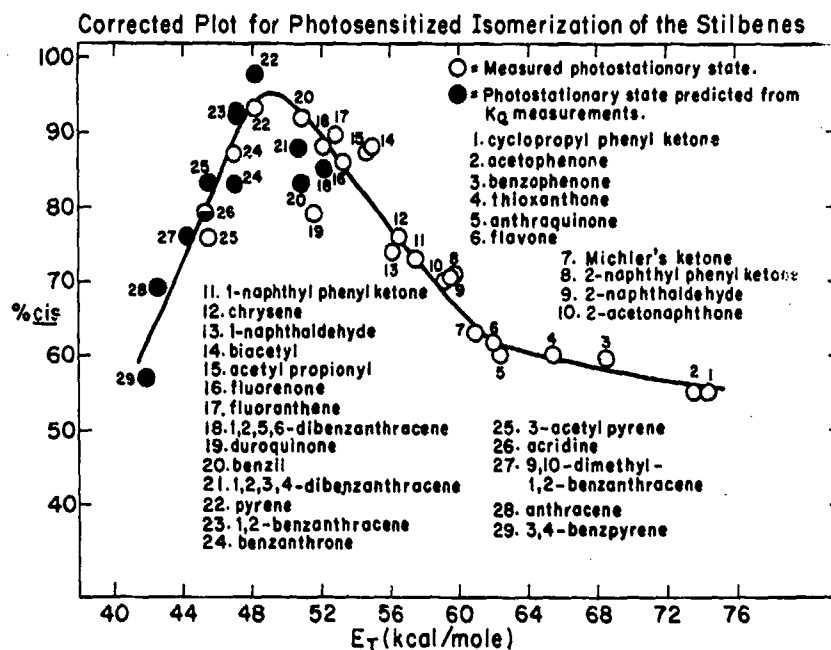
FIGURE 6.

As is indicated, the excitation energy of *cis*-stilbene is substantially higher than that of the *trans* isomer so the simple theory predicts that with low energy sensitizers, the photostationary states should contain too little *trans*-stilbene to be readily detectable. Figure 6 shows that the first parts of the prediction are correct. High-energy sensitizers do give essentially a common result, and decreasing the sensitizer energy below that of *cis*-stilbene produces a trend in the *cis*-rich direction. However, when the excitation energy of *trans*-stilbene is approached and passed, the trend is actually reversed and mixtures become more *trans*-rich. In attempting to explain this phenomenon we have decided that we must be observing unexpectedly high reactivity of *cis* stilbene as an energy acceptor with low-energy sensitizers. Our explanation is that this molecule is able to pass directly to a twisted state that has a lower energy content than any that can be reached by electronic excitation while maintaining the same geometry as is found in the ground state of the molecule. We call this geometrical accommodation of the energy acceptor "nonvertical excitation".

$$^3S_1 + \underset{H}{\overset{\phi}{C}}=\underset{H}{\overset{\phi}{C}} \longrightarrow {}^1S_0 + \underset{H}{\overset{\phi}{C}}-\underset{\phi}{\overset{H}{C}}$$
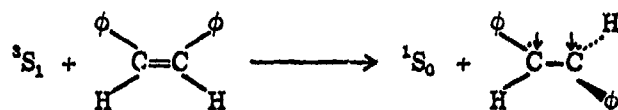
We were at first reluctant to invoke the concept of nonvertical excitation because it does not occur during light absorption. This is an application of the so-called Franck-Condon principle. However, there is really no reason to expect the Franck-Condon principle to hold for a process such as energy transfer. This is actually a simple form of a bimolecular reaction and does not involve any necessary interaction with the radiation field. Having once convinced ourselves that we needed a theory, we have since put it to use and discovered a number of unexpected new photosensitized reactions.

COMMON PHOTOCHEMICAL REACTIONS

Photochemical reactions can be roughly grouped into the following five classes:

1. Fragmentation
2. Ionization
3. Isomerization
4. Cycloaddition
5. Atom abstraction

The first three are unimolecular processes, in which an excited molecule either breaks into fragments or undergoes internal rearrangement. Reactions 4 and 5 involve attack of an excited molecule on some other species. I will present a few, nearly randomly chosen, examples of the various types.

Fragmentation Reactions. Figure 7 shows a number of representa-

$$CH_3COCH_3 \xrightarrow{h\nu} CH_3\dot{C}O + \cdot CH_3$$

$$R-N=N-R \xrightarrow{h\nu} 2R\cdot + N_2$$
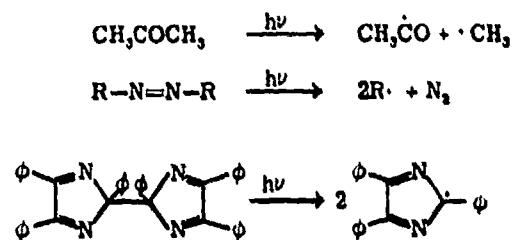
FIGURE 7. Formation of free radicals

tive fragmentation reactions. One of the oldest known photochemical reactions is cleavage to form free radicals. The process has been widely used to effect controlled initiation of chain reactions in which free radicals play a key role. For example, polymerization of butadiene or styrene to form high polymers, which are the base of synthetic elastomers and common plastic, can be initiated by radicals formed by photolysis of azo compounds $(R-N=N-R)$ or peroxides $(R-O-O-R)$.

The last of the radical-producing reactions shown in Figure 7 has an entirely different application. The lophyl radical
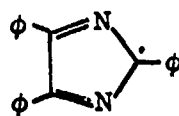
is highly colored whereas the parent, dimeric molecule does not absorb visible light. Irradiation of a material containing the dimer with ultra-violet light, which is absorbed, leads to photolytic production of the colored radical. The reaction is an example of *photochromism*, a process in which exposure of a material to light causes it to change color. The lophyl radicals normally combine to regenerate the parent compound; so the system is called a reversible photochromic system. Reversible photochromism is the subject of intensive study in many laboratories at the present time, because the effects have considerable potential value for information storage and relay and in development of autoprotective equipment, such as photochromic windshields for automobiles and airplanes. Another interesting application that I have heard proposed is the use of photochromic inks that would cause the girls in certain magazines, especially popular with males, to blush when the pages are opened and exposed to light. Unfortunately, the reversibility of the photochromic system is not perfect. The lophyl radicals undergo reactions other than dimerization, so a few are permanently lost in each light-dark cycle.

Formation of Ions. The reaction shown in Figure 8 is another kind of dissociative reaction, in which ions are produced. Photolytic ionization seems to be less common than photodissociation to form radicals; this may be only an artifact of the choice of systems for study. The example shown, photoionization of triarylmethane leu-

conitriles, is of some interest in photochromism because the tri-arylcarbonium ions (Ar³C⁺) are highly colored.

Figure 9 shows another kind of ionization reaction, in which an electron is ejected by the photoexcited species. The second example
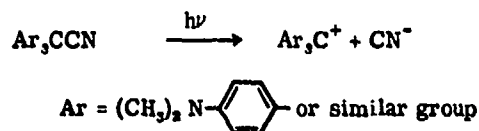
$$Ar_3CCN \xrightarrow{h\nu} Ar_3C^+ + CN^-$$
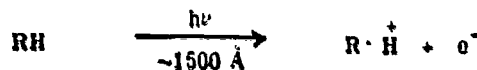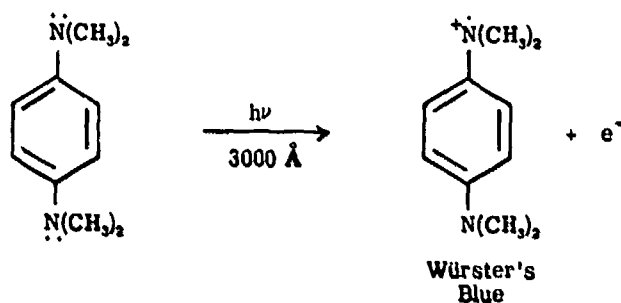
$$Ar = (CH_3)_2 N-\text{⟨benzene ring⟩}- \text{or similar group}$$

FIGURE 8. Formation of ions

Würster's
Blue

$$RH \xrightarrow[\sim 1500 \text{ Å}]{h\nu} R \cdot \overset{+}{H} + e^-$$

FIGURE 9. Ionization.

is probably a very general type of reaction. When irradiated with light of wavelengths shorter than about 1600 Å, most substances will probably yield photoelectrons. However, studies of this kind are not common, because light of wavelength shorter than 2000 Å is absorbed strongly by oxygen, necessitating the use of high vacuum equipment. Obviously "vacuum ultraviolet" photochemistry will be much more important in the everyday life of space travelers than to earthbound people. The ionization reaction shown at the top of Figure 9 occurs in solution under irradiation with near ultra-violet

light. The reaction is easily observed because the caton radical has an intense blue color. Despite its apparnt simplicity, photionization of Würster's base is rather complex. A quantum of 3000 Å light does not have enough energy to ionize a gaseous molecule of the base. Ionization probably occurs in some condensed media because the electron and cation can be stabilized by interaction with their environments. It is also likely that ionization results from the interaction of pairs of excited molecules, which results in concentration of all the excitation energy in one molecule.

Isomerization Reactions. Figure 10 shows a couple of interesting
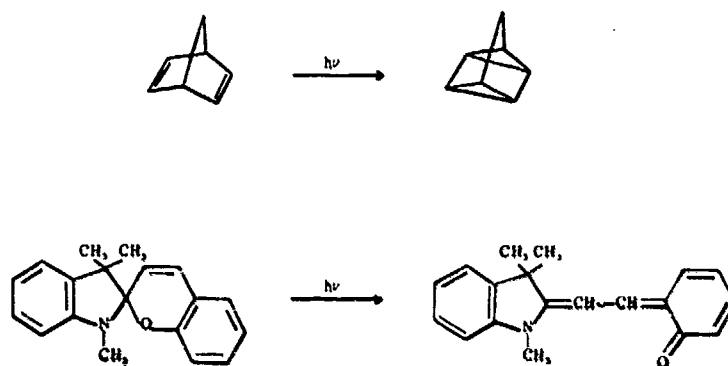
FIGURE 10. Isomerization

isomerization reactions. The first is of interest because the photoproduct has a much higher energy content than the starting material. Furthermore, the reaction can be carried out using sensitizers that absorb visible and near ultraviolet light. If we had a smooth way to catalyze the thermal reversion reaction, the system might be of some interest for storage of solar energy. However, I feel that the time is not yet ripe to fasten on a particular energy-storing system. Many others will be discovered as photochemistry forges ahead, and in another ten years the number of potential energy-storage and release systems may be increased tenfold. The second reaction shown in Figure 10 is another photochromic system that has received a good deal of attention recently. The colorless form is a member of a class of materials known as spiropyrans. The colored isomers can also be produced by heat, so the materials are thermochromic as well as photochromic.

Cycloaddition Reactions. Figure 11 shows a couple of examples of photoinduced cycloaddition reactions. The first example, dimerization of cyclopentenane, occurs under direct irradiation and by the sensitized technique. The sensitized reactions are believed to involve triplets; consequently, the fact that identical mixtures of the two products are formed in the direct and sensitized reactions implies that intersystem crossing occurs before the chemical action when light is absorbed directly. However, the relative amounts of the two
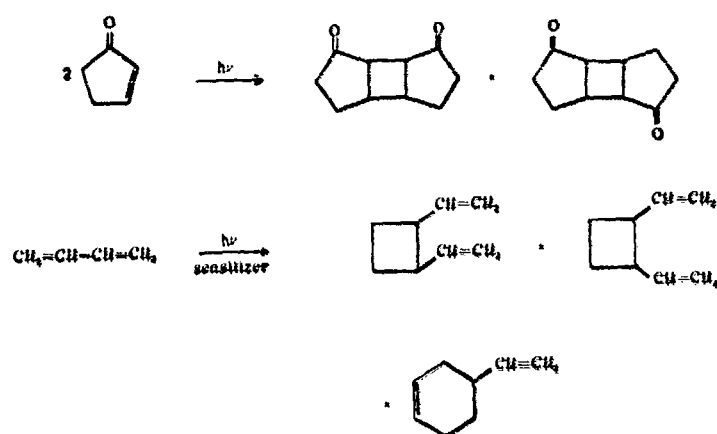
FIGURE 11. Cycloaddition.

products can be altered by changing the solvent in which the reaction is carried out. This simple mechanism for control of the course of a photoreaction was somewhat surprising to photochemists, although the phenomenon is very familiar from study of the dynamics of ordinary thermal reactions. There has existed a kind of mystique that goes something as follows: The specific rate constants for reactions of excited states are very large* and fast reactions are often both unselective and relatively insensitive to environmental influences. Other examples of solvent effects on the course of photoreactions have come to light within the last few years.

Dimerization of butadiene, the second reaction shown in Figure 12, is an example of a reaction that cannot be effected by direct exc'tation but goes smoothly using a variety of photosensitizers as

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* This must be so to allow chemical reaction to compete successfully with decay of the excited state back to the ground state.

Benzophenone

Hydrogen donor   Free radicals

$$2R \cdot \longrightarrow R-R$$
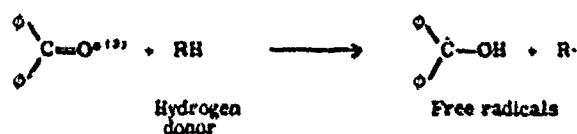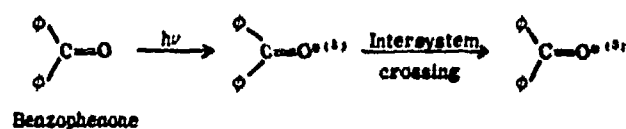
Figure 12. Atom Abstraction.

primary light absorbers. Furthermore, the relative amounts of the three products can be changed by varying the sensitizer. This fascinating effect is attributed to the existence of two, stereoisomeric triplet states of open-chain dienes.

cis triplet   trans triplet

The general lesson to be learned from this example is the necessity of recognizing that the geometry of an excited state may be significantly different from that of the same molecule in its ground state.

Atom Abstraction. The last type of reaction, atom abstraction, is illustrated in Figure 12. The name arises from a common mechanistic step, and not from the overall course of the reaction. The net reaction is usually an addition, but two steps are involved. The key is abstraction of an atom from some molecule by an excited species. The first step produces free radicals and the final products are formed by coupling of the radicals. Aldehydes and ketones, which contain carbonyl groups ($>C = O$), are most frequently the excited species and the atom abstracted is usually hydrogen. The reaction is probably the commonest method for photochemical production of free radicals in condensed media and is probably largely responsible for initiation of the oxidative reaction that leads to degradation of films and fibers under ordinary use conditions.

I have a feeling that the last part of this presentation may seem encyclopedic. However, if we could not claim some insight into the intimate details of photoreactions, we would have no grounds for hoping that controls necessary for future use of photoreactions as gears in chemically-based systems could be devised.

ACKNOWLEDGMENT

XI. Some Properties of the Liquid State

STUART A. RICE

A. Linear Transport Phenomena in Simple Liquids

1. INTRODUCTION

IF A TEMPERATURE gradient is maintained across a sample of some substance, there is established a steady state heat flow corresponding to the transport of energy from the hotter to the colder side. This is but one of many possible transport phenomena which can occur. Simple models descriptive of transport phenomena in a dilute gas have existed for about one hundred years, and a complete kinetic theory based on the Boltzmann collision equation for about fifty years.[1] In contrast, serious attempts to construct a kinetic theory of liquids started only twenty years ago.[2] The reason for a lag in the development of a theory of liquids is easy to find. In a dilute gas, molecules move along linear trajectories which are infrequently interrupted by binary collisions. Thus, the dynamical evolution of the state of the gas may be described in terms of the properties of successive uncorrelated binary collisions, and since the two body problem is easily solved there is no difficulty in representing the steady state linear transport phenomena in terms of the intermolecular potential and other molecular properties. Now, in a liquid or dense gas every molecule is in continuous interaction with a large number of near neighbors. A consequence of this state of simul-

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taneous interaction is the disappearance of the binary collision as a uniquely defined dynamical event. Because we cannot use isolated binary collisions as a basis for dynamical calculations, and because of the strong correlations between molecular positions, the description of the non-equilibrium steady state in a liquid must employ a more sophisticated analysis of N-body dynamics than is necessary in the description of the properties of a gas. Indeed, it will be necessary to examine in a fundamental way the nature of irreversibility in an N molecule system, and to learn how the description of irreversibility is related to N-body dynamics.

The starting point for the discussion of transport phenomena is the description of the macroscopic dissipative processes in terms of the constraints which define the nonequilibrium state of the system, and a set of coefficients which measure the rate of dissipation. Dissipative processes arise from the transport of mass, momentum and energy. In each case there exists a phenomenological relationship between a flux and the force which is responsible for the flux. In the cases of energy and mass transport we have the Fourier and Fick equations,

$$q = \kappa \nabla T,$$

$$\frac{\partial T}{\partial t} = \kappa \nabla^2 T,$$

$$J_m = D \nabla C,$$

$$\frac{\partial c}{\partial t} = D \nabla^2 C, \tag{1}$$

with q and J_m the energy and mass fluxes, κ and D the coefficients of thermal conductivity and diffusion, T the temperature, and c the concentration of one of the two components in the medium wherein diffusion is occurring. In the case of momentum transport the stress tensor σ and the rate of strain $\dot{e}$ play primary roles. For a Newtonian fluid the principal shearing stresses are proportional to the corresponding rates of strain and

$$\sigma = [-p + (\phi - 2/3\eta) \nabla \cdot u] \, \iota + 2\eta \dot{e}, \tag{2}$$

with ϕ and η the coefficients of dilatational and shear viscosity, u

the fluid velocity, p the pressure, and 1 the unit tensor. The stress law (2) when introduced in the equation of motion of the fluid leads to the Navier-Stokes equation—the starting point for the study of fluid dynamics.

For the simple fluids considered in this lecture, Eqs. (1) and (2) provide an accurate representation of dissipative behavior. The coefficients κ, D, η and ϕ may be determined experimentally by a variety of methods based upon suitable solution of the appropriate differential equation. It is found that all of the transport coefficients vary when the temperature and density of the liquid are varied. It is observed that, at constant external pressure, D increases exponentially and η decreases exponentially as T is increased. Under the same conditions κ is much less sensitive to changes in temperature than are η and D. For most simple liquids D is of the order of 10^{-5} cm^2/sec, κ of the order of 10^{-4} cal/cm sec °C, and η of the order of 5×10^{-3} dyne sec/cm.^{2} The dilatational viscosity, ϕ, is partially responsible for the attenuation of sound in a liquid. In simple liquids, e.g. Ar, N$_2$, ϕ is of the same order of magnitude as η.

Suppose that, for some liquid, the several transport coefficients have been determined as a function of temperature and density. How can these data be interpreted in terms of molecular dynamics and the structure of the liqu.? The extant theories of transport phenomena, which deal with just such an analysis, may be conveniently grouped into four classes: (a) simple quasi-solid or quasi-gas models with many empirical parameters, (b) phenomenological analyses based upon the principle of corresponding states, (c) statistical mechanical theories starting from the rigorous Liouville equation but employing simplifying approximations, and (d) developments which lead to formally exact results, but which may be difficult to use for a practical calculation.[3] In the following we examine the Rice-Allnatt theory which is an example of class (c). Mathematical details of the derivations are available in the literature[4] and will not be repeated herein. We shall focus attention exclusively on the nature of the physical arguments, the validity of the approximations, and the implications of the theory in other contexts. Wherever possible we shall also examine the agreement between theory and experiment, and the relationship between the Rice-Allnatt theory and the approaches of class (d).

2. GENERAL COMMENTS

The development of a statistical theory of transport phenomena in liquids requires consideration of many problems, among which are:

(a) Analysis of the means by which the time reversible equations of classical and quantum mechanics, which are used to describe the motions of molecules, lead to the time irreversible flux equations displayed in Equation (1),

(b) Derivation of a suitable kinetic equation determining the time evolution and phase dependence of some ensemble probability distribution,

(c) Solution of the kinetic equation to obtain relationships between the macroscopic parameters η, ϕ, κ and D and the intermolecular potential, number density, temperature, etc.

The reader should recognize that the calculation of transport coefficients is, in fact, only a small part of the general problem of describing time dependent phenomena. Namely, it is concerned with that state of a fluid in which all time dependence resides in the local hydrodynamic flow velocity. The general problem also involves the description of those short lived processes whose time dependence is explicit. Such processes generally depend on the nature of the initial state, the history of the evolution of the system and other factors. They are called non-Markovian processes because of the dependence on history, etc. In the hydrodynamic regime the processes involved depend only on the instantaneous state of the system, and are thereby classified as Markovian. The asymptotic approach of the exact kinetic equations describing non-Markovian procsses to the Markovian equations of the hydrodynamic regime, with which we are concerned in this section, is discussed later.

The exact kinetic equations for a dense fluid can be displayed only in the most formal way at the present time. Consequently, their asymptotic Markovian form is unknown, and the forms of the equations derived to describe a dense fluid are based on a set of approximations which simultaneously define both an intuitive analysis of the nature of random processes and a simple physical description of the fundamental dynamical processes influencing the evolution of the state of the liquid.

The method of obtaining equations satisfied by the one- and two-

molecule distribution functions $f^{(1)}$ $(\Gamma_1; t)$, $f^{(2)}$ $(\Gamma_2; t)$, respectively, is essentially that of integrating the N-molecule distribution function $f^{(N)}$ $(\Gamma_N; t)$ over the sub-phase space of all the other molecules in the system. Now, $f^{(N)}$ satisfies the Liouvile equation, and is not known explicitly. Therefore, one may only obtain differential equations for $f^{(1)}$ and $f^{(2)}$ by integrating the Liouville equation term by term. The result is a coupled hierarchy of equations: i.e., the equation for $f^{(1)}$ also involves $f^{(2)}$, the equation for $f^{(2)}$ also involves $f^{(3)}$, and so on. It is necessary to truncate or otherwise rearrange this hierarchy at some point in order to obtain closed equations for $f^{(1)}$ and $f^{(2)}$.

For a classical fluid we describe the system of N structureless molecules in the volume V by use of the Hamiltonian equations of motion. These equations have some interesting general implications. Since there is one equation for each degree of freedom of the system, it fol'ows that the phase of the system at any instant is uniquely determined by the phase at any other instant. In accordance with the definition of a Markov random process, it follows that the phase of the system, Γ_N, may be regarded as a Markov process of a simple kind. (The transition probability is a δ-function, since the increment of the variable Γ_N has only one possible value for each time instant.) The kinetic equations for the reduced distribution functions $f^{(1)}$, $f^{(2)}$, . . . , are concerned with the random variables Γ_1 (1), Γ_2, (1,2), . . . , which are of smaller dimensionality. Now, it is well known that the projection of a Markov process of 6 N dimensions onto a space of smaller dimensionality (6, 12, . . . , dimensions) generally yields a random process of higher order. Thus, Γ_1 (1), Γ_2 (1,2), . . . will be non-Markovian processes of higher order. This general feature has been obtained in the analysis of Prigogine and co-workers.[5] They find that the stochastic interaction term has the form of a time-convolution over the history of the variable. The important result is that when the system has reached a stationary state, the kinetic equations reduce to Markovian form.

The problem of analyzing further the coupled hierarchy of kinetic equations has, therefore, two distinct features. Since an integration over the sub-phase space of (N-1) or (N-2) molecules leaves the equations completely reversible, the analysis used must, in the first place, single out the features (e.g. time scale) which make the equations irreversible. Secondly, it must provide a means of picking out

the Markovian features that the kinetic equations contain in the hydrodynamic regime. The introduction of irreversibility is not difficult; it is merely contingent upon the particular method by which the Markovian feature of the description is achieved. For some particularly simple systems, for example, a heavy particle in a linear chain of harmonically coupled particles,[6] the equations of motion may be solved exactly. It is then found that there is a time scale on which the system exhibits irreversible behaviour in the sense that the velocity autocorrelation function of the heavy particle tends to zero, etc. For a sufficiently long time, recurrences of the mechanical state occur, and complete reversibility is observed. Whether or not irreversible behaviour is observed is then linked to the time scale on which the system is observed. The level of detail with which the dynamics is described is also of importance. For, mechanical quantities which depend on the positions and momenta of all N particles in the system do not exhibit irreversible behaviour, even in the sense described. Of course, almost all macroscopic properties of a system depend on the averaged behaviour of only a small number of molecules, say pairs, triplets, ... It is these quantities which behave as described.

Unfortunately, there are very few systems for which the N-particle dynamics can be analyzed exactly. In addition, at present no systematic and analytic procedure for determining the quasi-Markovian behaviour of an evolving system is known. Thus, it is necessary to adopt approximate methods to extract from the N-body dynamics the features desired. It must be understood that these approximate procedures do not "introduce" irreversibility where a complete and correct analysis would not. Rather, the approximations used are intended to provide adequate solutions to the N-body problem in some time or space domain. The one we shall adopt is based on that first proposed by Kirkwood.[2]

Consider now the relationship between non-Markovian processes in the subphase spaces of one, two, . . . molecules, and the ultimate transition to a Markovian kinetic equation defined on these same subphase spaces. We wish to assert that an nth order process can be treated as an n-dimensional Markov process, the reduction being accomplished by grouping the states of the process into hyperstates. Each hyperstate in the Markov process contains information about the history of the system during the interval t_m to t_{m+1}.

Much of this information is superfluous for the evaluation of the distribution functions in the hydrodynamic regime, but the information needed is contained within the hyperstate. The method of reducing the hierarchy of coupled equations for the distribution functions is therefore, *a means of extracting the relevant information from the hyperstate.* The particular contribution to the theory made by Kirkwood, which we have already mentioned, is the hypothesis that the relevant information for present purposes is contained in the exact distribution function averaged over an interval of time τ. The average value for an interval τ on the fine-grained time scale is then associated with a single point on a coarse-grained time scale, and the process is known as coarse graining (in time*). The kinetic equations obtained in this way are, in principle, difference equations, but it turns out that the times during which changes become significant on a hydrodynamic scale are so long compared to the coarse-graining time that no significant error is introduced by treating the differences as differentials.

The introduction of irreversibility which must accompany the coarse graining is accomplished by the assumption that a time interval τ exists such that the dynamical behavior of the system during one interval is related in a simple statistical manner to the dynamical events of the previous interval. It may be shown that the statistical character of the relation is sufficient to render the process irreversible.

The statistical assumption, or ansatz, can be analyzed on the basis of an intuitive picture of the dynamics of liquid molecules. Consider first the Fokker-Planck equation describing the behavior of a Brownian particle.[4] This equation describes a stochastic process under conditions such that the transition probability (for the phase Γ_B of the Brownian particle) is that for a stationary Markov process. In turn, this may be shown to be the result of allowing the time resolution of the description of the Brownian particle to be sufficiently coarse that transient behavior associated with the approach to local equilibrium in the molecular motions cannot be resolved. Thus, the description of Brownian motion as a Markov process applies only to the discussion of processes taking place on a time

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* It is possible also to coarse grain in space; similar kinetic equations are obtained.

scale longer than some τ_c characteristic of the dynamical behavior of the liquid molecules. In the development of the theory τ_c is chosen using physical criteria such that the basic dynamical event (in this case molecular fluctuations) is statistically independent of prior events. Were this not the case, the transition probability connecting two dynamical states of the Brownian particle would not be Markovian.

The problem of Brownian motion is concerned with numerous small momentum transfers, or numerous small particle displacements. At the other extreme of behavior, where momentum transfers may frequently be large and where displacements may be large, is the dilute gas. Transport phenomena in a dilute gas are usually described by a one molecule distribution function, which satisfies a kinetic equation (Boltzmann equation) in which the effects of molecular interaction appear in the form of isolated binary collisions.[1] The rate of change of the distribution function is determined by the slow secular variations of $f^{(1)}$ due to streaming in phase space, on which are superimposed the effects of the binary collisions. On the average, a molcule moves a long distance (relative to its size or the range of the intermolecular forces) before undergoing an encounter. Although there is a large volume of phase space wherein there occur small angle deflections resulting from binary collisions, large angle deflections are also frequent. Indeed, large angle deflections are responsible for most of the transport of energy and momentum due to collisions. From the numerous studies of the derivation of the Boltzmann equation from the first principles of statistical mechanics it is found that the assumptions required to effect a derivation are basically three in number: the neglect of interactions of higher order than binary collisions, the condition of molecular chaos (i.e., the condition that every pair of colliding molecules is statistically independent prior to the collision), and the slow secular variation of $f^{(1)}$ in space. Of these conditions, only the molecular chaos is responsible for the irreversibility.

Of course, in a liquid both small and large momentum transfers occur. How can we describe the properties of this system? The meaning of molecular chaos in a dilute gas is that molecule 2 (which is due to collide with molecule 1) has approached molecule 1 from infinity and its distribution of possible velocities has not been affec' d by collisions with molecules which have recently collided with mole-

cule 1. In a dense fluid, molecule 1 may undergo a rigid core, i.e., strongly repulsive, collision with a second molecule which has for some time past been in the region of the first coordination shell of molecule 1. Thus, molecule 2 should already have an intimate statistical "knowledge" of molecule 1, and may indeed have undergone a rigid core collision with molecule 1 in the immediate past. However, if the quasi-Brownian motion produced in the molecules by the van der Waals part of the forces is sufficiently effective in causing molecule 2 to forget its previous experience, then successive rigid core collisions should satisfy the simple form of molecular chaos used.

It is possible to formulate a consistency condition on the passage from the non-Markovian to the Markovian description of the fluid. In a sense, the distribution function may be thought of as a vector in a continuous space whose components represent the occupation probabilities of the various states of the phase space. In the most general case, the probability of finding the set of states $(p^{(n)}, \{n\})$ depends on the past history of the system. There is, however, a limiting case for which the past can be ignored:[a]

If, no matter what the sequence $(p^{(n)}, \{n\}, t_1), p^{(n)}, \{n\}, t_2), \ldots$, is, we always end up with the same assignment of probabilities for being in each of the states of $(p^{(n)}, \{n\}, t)$, then the preceding sequence can have no influence on the transition.

This condition is used as follows: Let it be assumed that there exists a time interval τ such that the following dynamical event, defined in τ, defines a Markov process. The dynamical event consists of a strongly repulsive binary encounter followed by a quasi-Brownian motion of the pair of molecules in the fluctuating field of all the neighboring molecules. Because the destruction of correlations by the quasi-Brownian motion is efficient, successive strongly repulsive encounters are statistically independent. The compound dynamical event is, therefore, asserted to be independent of prior events of the same kind.[*][†]

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[*] Recent studies of neutron diffraction from liquid Ar confirm the accuracy of this hypothesis B. A. Dasamacharya and K. R. Rao, Phys. Rev. 137, A417 (1965).

[†] The dynamical events are, of course, the interaction of the molecule, pair of molecules, etc., under consideration, with their environment. Clearly, the phase of the molecule, pair etc., is not independent of the phase during a previous interval; it is the phase of the environment which is (assumed) independent of the phase during a previous interval.

If, no matter what the sequence $(p^{(n)}, \{n\}, t_1), p^{(n)}, \{n\}, t_2), \ldots$, is, we always end up with the same assignment of probabilities for being in each of the states of $(p^{(n)}, \{n\}, t)$, then it is necessary that the relaxation time for return to the states of $(p^{(n)}, \{n\}, t)$ be short relative to the time interval on which the fundamental dynamical event is defined. Thus, if it can be shown that the relaxation time for the return to local equilibrium is much shorter than the time between strongly repulsive binary encounters, the initiation of the dynamical event consisting of a strongly repulsive binary encounter followed by a quasi-Brownian motion always starts from the same distribution function. In this case the probabilities for being in each of the states of $(p^{(n)}, \{n\}, t)$ just define the distribution function, and the required condition is satisfied.

A very interesting and fundamental analysis of the role of coherence time in the statistical mechanics of irreversible processes has been given by Fano[9] using some ideas and techniques introduced by Zwanzig.[10] Fano shows that in the limit that the dynamical coherence between a subsystem and its surroundings (reservoir) is short lived, the effective interaction between reservoir and system is weak irrespective of the magnitude of the intermolecular potential. From Fano's analysis Hurt and Rice[11] have developed a formal coherence time expansion for the classical fluid, and show that:

(a) In the limit of short memory of dynamical coherence, the Rice-Allnatt kinetic equations (see following) are a valid description of steady state phenomena in the liquid.

(b) Despite the fact that the usual expansion parameters $\rho \sigma^3$ or ϵ/kT are not useful in the liquid, there does exist a qualitatively different expansion parameter, τ_c/τ where τ_c is the lifetime of dynamical correlations and τ is the time between dynamical events. The new parameter appears naturally because, when the surrounding medium has the property of propagating away or otherwise destroying dynamical correlations in the subsystem of interest, it is not pertinent to measure the strength of the interaction in terms relating to the spacing of the continuous spectrum of the Liouville operator of the surrounding medium. All that is pertinent in this case is the lifetime of dynamical correlations. For the case of a perturbation in momentum space, Rice and Allnatt have shown[4] that the lifetime of the dynamical correlation is an order of magni-

tude less than the time between dynamical events, thus justifying truncation of the coherence time expansion after terms in τ_c/τ.

(c) The fundamental hypothesis of time smoothing is a natural expression of the nature of the coherence time expansion.

We have already mentioned the Rice-Allnatt kinetic equations (see (a) and (b)). These were developed before the derivation of the coherence time expansion, using physical arguments with content substantially identical to the formal results of the coherence time analysis. For simplicity, we shall discuss the theory in intuitive terms.

The theory of irreversible phenomena in liquids developed by Rice and Allnatt was, in the first instance, relevant to a model monoatomic dense fluid in which the intermolecular potential has the form of a rigid core repulsion superimposed on an arbitrary soft potential. Subsequent analysis has shown that the extension of the model to include more realistic potentials presents no formal difficulty, provided that the repulsive potential is sufficiently short ranged.

What advantage results from separating the intermolecular potential into two parts and treating their effects separately? Quite simply, the difference in range and strength of the repulsive core and the soft potential allows the discussion of the molecular motion in terms of two time scales: one corresponds to the large momentum and energy transfers which occur during a strongly repulsive encounter, while the other corresponds to the frequent small momentum and energy transfers which occur during the quasi-Brownian motion of a molecule in the superimposed soft force field of all the molecules in its surroundings. The short range of the strongly repulsive core implies that the first class of encounters are of short duration, so that the probability that a molecule undergoes such encounters with two or more others simultaneously is sufficiently small that it may be neglected. The introduction of the idealized rigid core representation for this class of encounters may thus be regarded as a formal device for restricting consideration to binary encounters (i.e., rigid core encounters between not more than two molecules). It has the additional advantage of considerably simplifying the mathematical details of the solutions of the equations but, we believe, without significantly affecting the numerical results.

The Markovian property of the kinetic equations is introduced

into the analysis by the use of the Kirkwood hypothesis that a time interval τ exists such that the dynamical events occurring in one interval are independent of those in the preceding intervals. The dynamical event is identified as a rigid core encounter followed by erratic or quasi-Brownian motion in the fluctuating soft force field of the neighboring molecules. This identification is contingent upon the effectveness of the quasi-Brownian motion at causing the environment to forget the momentum with which a molecule was rebounded after the rigid core encounter. This in turn implies that the relaxation time for the equilibrium of the momentum due to the soft force alone is much shorter than that due to rigid core encounters alone. It may be shown that this physical statement is supported by detailed calculation of the appropriate relaxation times for the motion considered.

The introduction of irreversibility in the manner described leads to a set of integrodifferential equations describing the evolution of the coarse-grained singlet $f^{(1)}$ (1), doublet $f^{(2)}(1,2)$, etc., distribution functions. Details of the derivations may be found elsewhere.[4] Here we merely state that each of the kinetic equations involves, in the interaction term, a sum of repulsive short ranged and weak longer ranged scattering operators. For the singlet distribution function the short ranged scattering term is similar in form to the Enskog kernel in the kinetic theory of the rigid sphere fluid.[1] The weak interaction scattering is described by a weak coupling master operator which, when the friction is independent of momentum, reduces to a Fokker-Planck operator. Rice and Allnatt use the form involving the Fokker-Planck operator because the more general kinetic equation is so complex that solutions are difficult to obtain. It should be noted that the weak coupling part of the equation derived by the use of the Kirkwood hypothesis is identical with that derived by Prigogine.[5] The coherence time expansion of Hurt and Rice also leads to the Rice-Allnatt kinetic equation. Finally, using a functional integral approach which completely eliminates the use of the Kirkwood hypothesis, Popielawski and Rice[12] have derived the Rice-Allnatt kinetic equation and shown its relationship to a summed form of the Prigogine perturbation theory.

The Rice-Allnatt kinetic equations may be solved analytically when there are only small deviations from equilibrium. The solutions, which depend on the temperature gradient, velocity gradient,

etc., may then be used to compute the several transport coefficients. The results are:

(a) Thermal conductance:

$$\kappa = \kappa_k + \kappa_v(\sigma) + \kappa_v(R > \sigma). \tag{3}$$

$$\kappa_k = \frac{75k^2T}{32\,mg(\sigma)}\left[\frac{1 + (2\pi\rho\sigma^3/5)\,g(\sigma)}{\Omega^{(2,2)} + (45\,\zeta_8/16\rho mg(\sigma))}\right],$$

$$\Omega^{(2,2)} = (4\pi kT/m)^{1/2}\,\sigma^2,$$

$$\kappa_v(\sigma) = \frac{75k^2T}{32mg(\sigma)}\left[\left(\frac{2\pi\rho\sigma^3}{3}\right)C + \left(\frac{2\pi\rho\sigma^3 g(\sigma)}{5}\right)\left(\frac{32}{9\pi^{3/2}}\right)\right]$$

$$C = \frac{\left[1 + \dfrac{2\pi\rho\sigma^3}{5}g(\sigma)\right]}{\left[2\Omega^{(2,2)} + \dfrac{45\,\zeta_8}{16m\rho\,(g(\sigma))^2}\right]}\cdot\left[1 + \frac{\Omega^{(2,2)}\,g(\sigma)}{\Omega^{(2,2)} + \dfrac{45\,\zeta_8}{16m\rho g(\sigma)}}\right],$$

$$\kappa_v(R > \sigma) = \frac{\pi kT\rho^2}{3\zeta_8}\int_\sigma^\infty R^3\left(R\frac{du}{dR} - u\right)g(R)\frac{d}{dR}\left[\frac{\partial \ln g(R)}{\partial T}\right]dR$$

$$+ \frac{\pi kT\rho^2}{\zeta_8}\int_\sigma^\infty\left(R^2u - \frac{1}{3}R^3\frac{du}{dR}\right)\left(\frac{\partial g}{\partial T}\right)dR. \tag{4}$$

(b) Shear viscosity:

$$\eta = \eta_k + \sum_{i=1}^{3}\eta_v^{(i)}(\sigma) + \eta_v(R > \sigma). \tag{5}$$

$$\eta_k = \frac{5kT}{8g(\sigma)}\cdot\frac{[1 + (4\pi\rho\sigma^3 g(\sigma)/15)]}{[\Omega^{(2,2)} + (5\zeta_8/4\rho mg(\sigma))]},$$

$$\eta_v^{(1)}(\sigma) = \frac{5kT}{8g(\sigma)}\left(\frac{2\pi\rho\sigma^3}{15}\right)\left[1 + \frac{4\pi\rho\sigma^3}{15}g(\sigma)\right]D,$$

$$\eta_v^{(2)}(\sigma) = \frac{8\pi}{15}\frac{\rho^2\sigma^4 kTg(\sigma)}{\Omega^{(2,2)}},$$

$$\eta_v^{(3)}(\sigma) = -\frac{57}{70}\left(\frac{2\pi\rho\sigma^3}{15}\right)\rho g(\sigma)\zeta_8\,\psi_2(\sigma).$$

$$D = \left[\Omega^{(2,2)} + \frac{5\zeta_s}{8\rho m(g(\sigma))}\right]^{-1} \cdot \left[1 + \frac{4\Omega^{(2,2)}g(\sigma)}{4\Omega^{(2,2)} + \frac{5\zeta_s}{\rho m g(\sigma)}}\right],$$

$$\eta_v(R > \sigma) = \frac{\pi\rho^2\zeta_s}{15kT} \int_\sigma^\infty R^3 \frac{du}{dR} g(R) \psi_2(R) dR. \tag{6}$$

with the function $\psi_2(R)$ obtained as the solution to the differential equation

$$\frac{d}{dR}\left(R^2 g(R) \frac{d\psi_2}{dR}\right) - 6g(R)\psi_2 = R^3 \frac{dg}{dR}. \tag{7}$$

with boundary conditions

$$\lim_{R \to \infty} \psi_2(R) = 0.$$

$$\lim_{R \to \infty} R^2 g(R) \frac{d\psi_2}{dR} = 0. \tag{8}$$

(c) Bulk viscosity:*

$$\phi = \sum_{i=1}^3 \phi_v^{(i)}(\sigma) + \phi_v(R > \sigma). \tag{9}$$

$$\phi_v^{(1)} = 0.$$

$$\phi_v^{(2)} = \frac{5}{3}\eta_v^{(2)}.$$

$$\phi_v^{(3)}(\sigma) = \frac{2\pi\rho\sigma^3}{15} \cdot \rho g(\sigma) \zeta_s \left[\frac{4\psi_2(\sigma) - 35\psi_v(\sigma)}{42}\right],$$

$$\phi_v(R > \sigma) = \frac{\pi\rho^2\zeta_s}{9kT} \int_\sigma^\infty u^1(R)g(R)\psi_v(R)R^3dR. \tag{10}$$

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* There is no kinetic contribution to the bulk viscosity; in other words: $\phi_k = 0$.

(d) ion mobility:

$$\mu_+ = \frac{q}{\frac{8}{3}\rho\sigma^3 g(\sigma)\left(\frac{2\pi m m_i kT}{m+m_i}\right)^{1/2} + \zeta_S}, \tag{11}$$

where q is the charge on the ion, m_i the mass of the ion, and the appropriate value of ζ_S is for the ion-molecule core interaction.

(3) The friction coefficient ζ_S, which appears in all the preceding formulae, has not yet been computed with comparable accuracy. Three different theoretical estimates are:[5]

(i) $\zeta_S^2 = \frac{m\rho}{3}\int \nabla^2 u(R)g(R)d^3R,$ $\tag{12}$

(ii) $\zeta_S = -\frac{\rho}{12\pi^3}\left(\frac{\pi m}{kT}\right)^{1/2}\int k^3\,\tilde{u}^s(k)\tilde{G}(k)dk,$ $\tag{13}$

with

$$\tilde{u}^s(k) = \int u^s(R)\,e^{i\mathbf{k}\cdot\mathbf{R}}d^3R.$$

$$\tilde{G}(k) = \int [g(R) - 1]\,e^{i\mathbf{k}\cdot\mathbf{R}}d^3R,$$

(iii) $\zeta_S = \zeta_S^{(1)} + \zeta_S^{(2)}$

$\zeta_S^{(1)} = -\frac{8}{3}\sigma^2\rho g(\sigma)\,(\pi mkT)^{1/2}\,J^{(1)},$

$\zeta_S^{(2)} = -\frac{8}{3}\sigma^2\pi^2 g(\sigma)\frac{\rho^2}{(2\pi)^6}\,(\pi mkT)^{1/2}\,J^{(2)},$

$J^{(1)} = \frac{1}{4\pi\sigma^2 kT}\int_\sigma^\infty dk[k\sigma\cos(k\sigma) - \sin(k\sigma)]\tilde{u}^s(k)$

$$J^{(2)} = \frac{1}{4\pi\sigma^2 kT}\int_\sigma^\infty dk\,\ell\frac{\boldsymbol{\ell}\cdot\mathbf{k}}{|\mathbf{3k}|}[l\sigma\cos(l\sigma) - \sin(l\sigma)]\,\tilde{u}^s(k)\tilde{G}(l)\tilde{G}(|\mathbf{k}-\boldsymbol{\ell}|).$$

$$\tag{14}$$

A discussion of these formulae is deferred to Section 3.

In Eqs. (3) — (14), m is the mass of a molecule, σ the hard core diameter of a molecule, ρ the number density of the liquid, $g(\sigma)$ the

pair correlation function when two molecules are in contact at $R = \sigma$ and $g(R)$ the pair correlation function for a molecular separation R, ζ_S the friction coefficient arising from the autocorrelation of the soft (longranged) component of the force acting on a molecule, u the intermolecular pair potential, $u^s(k)$ the Fourier transform of the soft part of the intermolecular potential and $G(k)$ the Fourier transform of $(g(R) - 1)$.

3. COMPARISON OF THEORY AND EXPERIMENT

We now consider how the predictions embodied in Eqs. (3)— (14) agree with the available data. First, however, we may ask which data are available. In the case of simple liquids the most commonly measured transport property is the shear viscosity.[3] Fewer measurements of the thermal conductivity have been made, and the self-diffusion coefficient is known only for a very small number of simple liquids: Naghizadeh and Rice[13] studied Argon, Krypton, Xenon and Methane, while Cini-Castagnoli[14] reported one measurement of D for liquid Argon at 84.5° K (which is in fairly good agreement with the measurements of Naghizadeh and Rice), and a few measurements of D for liquid CO[15]. The diffusion coefficient of liquid CH_4 has also been deduced from spin echo measurements.[16] There are still fewer experimental determinations of the bulk viscosity. Studies of Ar and N_2 have been reported by Naugle and Squire.[17] The mobilities of ions in liquid Ar, Kr and Xe have been studied experimentally by Davis, Rice and Meyer,[18] and by Henson[19] (Ar), who has also measured the mobility of positive ions in liquid Nitrogen. Henson's results for liquid Ar are in agreement with those of Davis, Rice and Meyer.

The Rice-Allnatt theory predicts that, at constant density, the shear viscosity is little affected by changes in temperature, and Lowry, Rice and Gray[20] have shown that, in view of the sensitivity of the theory to the imperfectly known radial distribution function, this prediction is in agreement with Zhdanova's results for liquid Argon[21] (see Table I).

The temperature dependence of the thermal conductivity deviates slightly from linearity in the temperature region up to the critical temperature, in the direction such that it decreases with increasing temperature. We also note that the magnitude, and the pressure and

TABLE I
Shear Viscosity of Liquid Argon (millipoise)

State	90° K 1.5 atm	128° K 50 atm	133.5° K 100 atm	185.5° K 500 atm
η (obs)	2.39	0.855	0.843	0.869
η (calc)[a]	1.41	0.692	0.701	0.874
η (calc)[b]	1.67	0.817	0.821	1.000
η (obs) T η (obs)$_{128}$°	——	1.00	1.01	1.04
η (calc) T η (calc)$_{128}$°	——	1.00	1.005	1.06

a Rice-Allnatt theory
b Wei-Davis modification of Rice-Allnatt theory.

the temperature dependences of κ, all decrease in the following or-
der:[22] CH_4, Ar, Kr, Xe. Above 100 atm the coefficient of thermal con-
ductivity seems to increase almost linearly with the pressure and we
mention that, according to Ikenberry and Rice[22] $(\partial\kappa/\partial p)_T$ for CH_4
is considerably larger than for Ar, Kr and Xe. These investigators
tested the Rice-Allnatt theory against their experimental results
and found good agreement ($\sim 10\%$) (See Table II).

TABLE II
Thermal Conductivity of Liquid Argon
(Units of κ are 10^{-5} cal/sec. cm°K)

State	128° K 50 atm	133.5° K 100 atm	185.5° K 500 atm
κ(obs)	18.9	18.6	18.7
κ(calc)[a]	16.9	15.9	17.0
κ(calc)[b]	12.5	14.0	17.0

a Rice-Allnatt theory.
b Wei-Davis modification of Rice-Allnatt theory.

Examination of the Rice-Allnatt theory shows that one of the
major theoretical problems is the determination of the self-diffusion
coefficient, not only because diffusion is a purely kinetic phenome-
non, but also because the Rice-Allnatt determination of the other
transport properties depends strongly on the value of the friction
coefficient, i.e., on D. The experimental results of Naghizadeh and
Rice[13] fit very well a linear relationship for the isobaric temperature
dependence of the logarithm of D. Naghizadeh and Rice also ob-
serve that the self-diffusion coefficient decreases exponentially with
increasing pressure at constant temperature and that, in contrast to

the thermal conductivity, $(\partial D/\partial p)_T$ is much smaller for CH_4 than for Ar, Kr, and Xe. The calculations displayed in Table III show that none of the theoretical descriptions of the friction coefficient is completely adequate. The source of error is easily traced to an inadequate analysis of the autocorrelation function. Eq. (12) corresponds to a Gaussian autocorrelation function, which cannot reproduce cage effects in the liquid. The linear trajectory approximation, either without correlation between soft and hard forces (Eq. (13)) or with such correlation (Eq. (14))[23] is better in predicting the magnitude of D, but the predicted temperature dependence of D is not completely satisfactory, although it is not badly in error. There is also one simple model, the dense square-well fluid,[24] which provides a useful zeroth order approximation to the behavior of real fluids. Davis, Rice and Sengers used this model to study the friction coefficient.[24] Although the magnitude of the predicted diffusion coefficient is too small by $\sim 30\%$ when one assumes there is an exponential decay of the velocity autocorrelation function, the predicted temperature dependence of D is excellent (see Table III).

TABLE III
Self-Diffusion Coefficient for Liquid Argon
(Units of D are 10^{-5} cm^2 sec^{-1})

	84° K	90° K	100° K
D (obs) [*]	1.84	2.35	3.45
D (calc)[a]	1.43	1.80	2.25
D (calc)[b]	3.91	4.11	——
D (calc)[c]	2.25	2.49	——
D (calc)[d]	2.80	3.25	3.85
D (calc)[e]	2.46	2.70	3.34

[*] Naghizadeh and Rice

a. Square Well, exponentially decaying correlation function.

b. Small Step Diffusion Theory (Eq. (12)).

c. Small Step, isotope separation data (Eq. (12)).

d. Linear trajectory theory with no cross correlations (Eq. (13)).

e. Linear trajectory theory with inclusion of cross correlations (Eq. (14)).

Buttressing the reality of this agreement, Davis and Luks[25] have recently used the square-well model with great success for extensive computations of all the transport properties of liquid Ar, Kr and Xe. Although the square-well potential is certainly unrealistic, it does have the major features of a realistic pair potential. Concerning the pair potential and radial distribution function, it is interesting to

note that the poor agreement between experimental data and the Rice and Kirkwood small step-diffusion theory[26] can be considerably improved when the average Laplacian of the intermolecular potential is evaluated from isotope separation data, as suggested by Friedman and Steel[27] and by Boato, Casanova and Levi[28] (see Table III).

We now turn briefly to the study of ion mobility in simple liquids. To date, the literature on this subject is almost entirely limited to the experimental and theoretical study by Davis, Rice and Meyer[18] on the mobilities of positive and negative ions in liquid Ar, Kr, and Xe. We therefore refer the interested reader to the details presented in the original papers, and also to the monograph by Rice and Gray.[4] For our purposes it is sufficient to mention that the experimental data indicate that the mobility varies linearly, but very smoothly, with the external pressure, while the logarithmic dependence of the product μT can be represented adequately by the Einstein relation. The magnitude, the pressure dependence and the temperature dependence of the positive ion mobility in liquid Ar, Kr, and Xe can be quantitatively accounted for by the Rice-Allnatt theory, and the agreement with experiment is very satisfactory if the positive ions are Ar_2^+, Kr_2^+ or Xe_2^+, while it is much poorer if a different ionic species (say, Ar^+) is postulated. On the other hand, the study of negative ions is much more difficult because of impurity effects. Indeed, Davis, Rice and Meyer interpret their mobility data in terms of the properties of the O_2^- ion, and, if it may be assumed that the negative charge carriers in liquid Ar, Kr and Xe are effectively O_2^- ions, the Rice-Allnatt theory is again seen to provide an adequate representation of the observations.

Finally, consider the coefficient of bulk viscosity. ϕ can be determined from the excess ultrasonic attenuation (excess over that due to shear viscosity and thermal conductivity) and the only available data are for liquid Argon and liquid Nitrogen. These measurements demonstrate that: (a) in the high temperature region, the Rice-Allnatt theory gives a quantitative description of the density dependence and magnitude of the bulk viscosity, (b) at high density and low temperature the absolute magnitude of ϕ is predicted to within ~50%, (c) the predicted ratio $(\phi/\eta) = 1.3$ is within ~20% of the observed value of (ϕ/η). The experimental data also shows that (ϕ/η) decreases as ρ increases. No calculation of the dependence

of ϕ on density has yet been made. At the lowest temperature and highest density studied, the theory is clearly in disagreement with experiment. Gray and Rice assign the error to inadequacy of the available radial distribution function. In view of the successes of the theory under conditions where it is reasonable to believe that the distribution functions are not too bad, this conclusion seems valid. Indeed, a general examination of the agreement between theory and experiment convinces us that in all cases thus far examined the major contribution to the observed disagreement arises from the inadequacy of the available radial distribution function. It is clear that complete and definitive testing of the Rice-Allnatt theory awaits the determination of very accurate equilibrium distribution functions and potential functions. The presently available agreement between theory and observation suggests (but does not prove) that the Rice-Allnatt theory is a good first order theory of transport in liquids.

4. DISCUSSION

Since the space available is limited there are only a few more remarks which I can make. First, I wish to mention that the theory of the autocorrelation function, which is the most important part of the theory of the friction constant, has been advanced in recent studies. Using a memory function formalism Berne, Boon and Rice[29] have shown how the autocorrelation function and power spectrum of the linear momentum, including the effects of recoil, can be simply interpreted. This same formalism can be used to generate a linear trajectory analysis of the transport coefficients,[30] and a description of the relationship between initial correlations, dynamical memory, and the generalized Prigogine collison operator.[31]

Second, since I have several times alluded to the Prigogine theory, and used it as a bench mark against which approximate analyses were measured, it seems appropriate to give the following very brief description. The original analysis of Priogogine and co-workers used a Fourier decomposition of the N-body distribution function, $f^{(N)}$, and the classification of terms which appear in the decomposition according to powers to t, N/V, and λ where λ is the coupling constant of the intermolecular potential energy. In this formalism the Liouville equation takes a form which describes the transi-

tions between different distributions of wave vectors; the transitions are generated by the interactions between the molecules, and the distributions of wave vectors are the respective Fourier space representations of the distribution function. The equations are naturally ordered in a sequence which counts the number of non-zero wave vectors. To evaluate the terms in this representation, Prigogine and Balescu have invented a diagrammatic notation.[5] Later, by the use of operator techniques to solve the Liouville equation, Prigogine and Resibois have shown that[5]

$$\frac{\partial \rho_0(t)}{\partial t} = \mathcal{D}_0 \ (t, \rho_{\{k\}}^{(0)}) + \int_0^t G_{00} \ (t - t') \, \rho_0 \, (t') \, dt' \tag{15}$$

In (15) $\rho_{\{k\}}$ is the $\{k\}$ Fourier component of the distribution function. The exact master equation (15) describes the time evolution of ρ_0 (which is the velocity distribution function) for all time. The structure of the equation is very simple: the inhomogeneous term $\mathcal{D}_0$ gives the contribution, at time t, of the initially excited Fourier components which through interaction decay towards a state with no correlations; the second term has a nonlocal structure so that $\partial \rho_0 / \partial t$ depends upon $\rho_0(t')$ for times $t' < t$, and is representative of the fact that ρ_0 in general changes during a collision. Note that all the effects of the initial correlations and initial conditions appear in the term $\mathcal{D}_0$. A kinetic equation for the nondiagonal Fourier components, $\rho_{\{k\}}(t)$ may also be obtained. For this and other applications the reader is referred to the monograph by Prigogine and the original literature.

The first use to which the general master equation can be put is the examination of kinetic equations in the limit $t \to \infty$. In the transition to the limit it is seen that not only do all the effects contained in $\mathcal{D}_0$ wash out, but also that all effects arising from the finite duration of the binary encounters still do not prevent the kinetic equation being Markovian in the limit $t \to \infty$. For example, the phenomenological transport coefficients involve only the asymptotic cross sections, and no terms appear which are related to the duration of an encounter, except in the case of the bulk viscosity.

The appearance of the time convolution in the generalized master equation specifically includes contributions to $\partial \rho_0 (t)/\partial t$ from $\rho_0(t')$ for $t' \leq t$, with a weight $G_{00} (t-t')$. The behavior of $G_{00} (t-t')$

is determined by the intermolecular forces, density, etc., but not by the initial state of the system. Thus, the role of the convolution as such will only be important for times of the order of the interaction time τ_c, giving rise to transient effects. For $t \gg \tau_c$, the velocity distribution will vary only a little during τ_c, and the operator

$$\int_0^t G_{00} \, (t - t') \, dt'$$

will be approximately independent of t. The kinetic equation will then have the Markovian form

$$\frac{\partial \rho_0}{\partial t} = \mathscr{G}_{00} \, \rho_0, \tag{31}$$

where the operator $\mathscr{G}_{00}$ is given by

$$\mathscr{G}_{00} = \int_0^\infty G_{00} \, (t - t') \, dt'. \tag{32}$$

At this stage, correlations over distances of the order $(p/m) \, \tau_c$ will have been destroyed and the system will be evolving in the kinetic regime.

This description bears a considerable resemblance to the role which was assigned to the time coarse graining in the Kirkwood analysis, where, in order to develop an explicit representation of $\mathscr{G}_{00}$, a mechanism for the interactions was proposed. Equally important is the difference between the coarse graining proposed by Kirkwood and the way in which $\mathscr{G}_{00}$ is reached. The simple form of coarse graining involves an unweighted time average, while $\mathscr{G}_{00}$ is the result of a complex weighting determined by the nature of the interaction.

The method used to obtain a master equation for the velocity distribution function may also be used, with slight extension, to describe the time evolution of the molecular correlations. Again, a non-local equation is found to hold for all t, reducing to a Markovian equation in the limit $t \to \infty$.

In brief, then:

(a) The general kinetic equation is non-Markovian.

(b) For times long compared to the duration of an encounter

(and to other characteristic times in more general cases) it reduces to a Markovian equation, which may or may not require correction for effects arising from the finite duration of an encounter.

(c) For quasi-stationary situations, only the asymptotic form of the diagonal fragment enters the collision operator.

Thus, the form of the kinetic equation depends on the type of process being described and on the time scale of interest.

It is interesting to compare the Kirkwood coarse-graining hypothesis with the neglect of the initial correlations described by $\mathcal{D}_0$ and the transition to Markovian behavior. First, it should be noted that $\mathcal{D}_0$ tends to zero as t increases for the reason that correlations of finite extent in the initial state can only interact for a finite time. Indeed, if the range of the correlations is of molecular dimensions, then the lifetime of the initial correlations is of the order of an interaction time.[*] Second, the effect of the non-Markovian kernel is to connect the distribution function to itself over times of the order of the duration of an encounter. Now the fundamental idea involved in the use of coarse graining is that the dynamical event in τ is independent of prior dynamical events. This means that on the time scale chosen, $\mathcal{D}_0$ must vanish and that the time integral involving $G_{00}(t)$ must approach a limit independent of τ. In section 2 we remarked that if the distribution function returned to the form characteristic of the local environment on a time scale short compared to τ, then the process defined by time smoothing became a Markov process. Moreover, for the case of a perturbation in momentum space, Rice and Allnatt have shown that the single kinetic equation is consistent with this condition. It is clear that, in effect, the calculation of the relaxation time for a perturbation in momentum space is equivalent to the calculation of the lifetime of the correlations in a specified initial state. The consistency in this regard shows that $\mathcal{D}_0$ can be neglected under the conditions described by the Rice-Allnatt equation and that the use of time coarse graining does lead, as expected, to $\mathcal{D}_0 = 0$. Of course this is shown only for a special case, but the physical description is clear enough that the argument can be extended. For some states $\mathcal{D}_0$

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[*] The lifetime of the correlations is, in fact, of the order of a relaxation time. Nevertheless, the destruction term vanishes in an interaction time.

cannot be neglected (spin-echo experiment) and each situation must be separately analyzed. It is safe to conclude however, for the liquids we have considered, that coarse graining does lead to an equation from which all information about the initial correlations has been removed.

Now, both the Hurt-Rice and Popielawski-Rice analyses show that the Rice-Allnatt equation is the first term in a well defined approximation scheme in which:

a) The dynamical effects of a strong, short ranged repulsion between the molecules is approximated by successive uncorrelated quasi-binary encounters,

b) The dynamical effects of a weak, longer ranged attraction are included to all orders of perturbation theory,

c) The effects of the weak interaction on the dynamics of the quasi-binary encounter are neglected.

If the Rice-Allnatt equation is compared with the Prigogine theory, what is included and what is left out? The derivation of the Rice-Allnat equation shows that it includes contributions from:

a) all terms leading to the weak master equation,

b) all terms in the binary collision expansion corresponding to uncorrelated successive binary collisions.

Terms not mentioned under (a) or (b) are not totally neglected. The use of the local equilibrium approximation (and the explicit refusal to expand $f^{(2)}$ in a power series in ρ) is equivalent to the assumption that all remaining terms in the expansion are replaced by the corresponding equilibrium terms. Thus, the dynamics of the pair of molecules is influenced by the stationary field of the surrounding molecules, but the reaction of the surrounding molecules to the motion of the pair of molecules, and the instantaneous effect of the motion of the surroundings on the pair of molecules are neglected. The local equilibrium approximation can be thought of as replacing the N body dynamical problem by a two body problem with boundary conditions specified in terms of the equilibrium distribution of the other molecules of the system.

We conclude that there is both theoretical and experimental basis for the belief that the Rice-Allnatt kinetic equation is a reasonable zeroth order description of a simple liquid.

ACKNOWLEDGMENTS

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B. Studies of the Electronic States of Simple Liquids

1. INTRODUCTION

Considerable effort has been devoted to studies of the electronic states of free molecules and of crystals. Consider first the spectrum of bound states. Studies of dilute gases, from which information about the free molecule is deduced, are simplified by the absence of inter-molecular interactions and hence of correlations between the positions of the molecules. Thus, any one molecule may be regarded as isolated except for occasional binary collisions. Binary collisions, which decrease in frequency as the gas density is decreased, lead to a broadening and shift of the stationary states of the gas, and from this alteration of the spectrum there can be deduced information about the intermolecular potential.[1] A different simplification of description is possible in the case of crystalline solids. The long range order of a crystalline lattice is a consequence of strict trans-

lational symmetry. In turn, the existence of strict translational symmetry, as well as the presence of other geometric symmetries within the unit cell, permits the use of a description in which independent collective coordinates are fundamental to the representation. These collective coordinates, of course, correspond to the exciton states of the crystal. Interaction between the exciton states and lattice vibrations leads to some alteration of the spectrum, and useful information may thereby be obtained.[2]

Similar considerations may be used to describe the conduction electron states. In a gas, simple kinetic theory, assuming binary collisions, is sufficiently accurate. In a crystal, an electron may be described as interacting with collective vibrational modes, the phonons. In both cases, the scattering leading to finite electron mobility is readily interpreted in terms of a simple interaction potential and geometric configuration.

Now, a liquid has short range structural order but no long range structural order. Because of the strong interactions between the molecules in a liquid, approximations suitable to the description of a gas are not useful. Furthermore, the lack of a simple geometric symmetry in the short range order of the liquid makes it necessary that the concepts of the exciton theory of crystals and the band theory of conduction be extensively modified before they can be applied to the description of a liquid. It is my opinion that the development of a physically realistic and incisive interpretation of the electronic states of liquids and of other disordered systems is one of the most interesting and most challenging of contemporary scientific problems. One outgrowth of this contention is a program, initiated a few years ago at the University of Chicago, to study the electronic states of simple liquids. This report presents a very short resumé of some of our work on the electronic properties of simple monoatomic dielectric liquids, together with some comments on what we now understand, do not understand, and where new concepts and constructs are needed.

2. EXCESS ELECTRON STATES IN MONOATOMIC DIELECTRIC LIQUIDS

a. Free Electron States

We consider, first, the properties of an excess electron in a liquid

composed of neutral, closed shell atoms (for example, Helium, Argon, Krypton). Because the interaction between an electron and a neutral atom is both weaker and of shorter range than the interaction between an electron and an ion, it is tempting to suppose that an excess electron in a simple liquid behaves very much like a free electron. Of course, the fact the electron-atom interaction is non-zero suggests that scattering phenomena cannot be totally neglected, and that the free electron spectrum of states will be perturbed by the presence of fluid atoms.

Given presently available technology and the very low level at which excess electrons may be introduced into a simple dielectric liquid, most of the available experimental data on the properties of excess electrons are obtained from mobility measurements. Referring the reader elsewhere for detailed descriptions of the experimental techniques,[3] we display in Figs. 1, 2, and 3 the results of mobility measurements made in a time of flight instrument at very low electron concentrations (10^5 cm^{-3} ~ 10^{-18} molar). These data
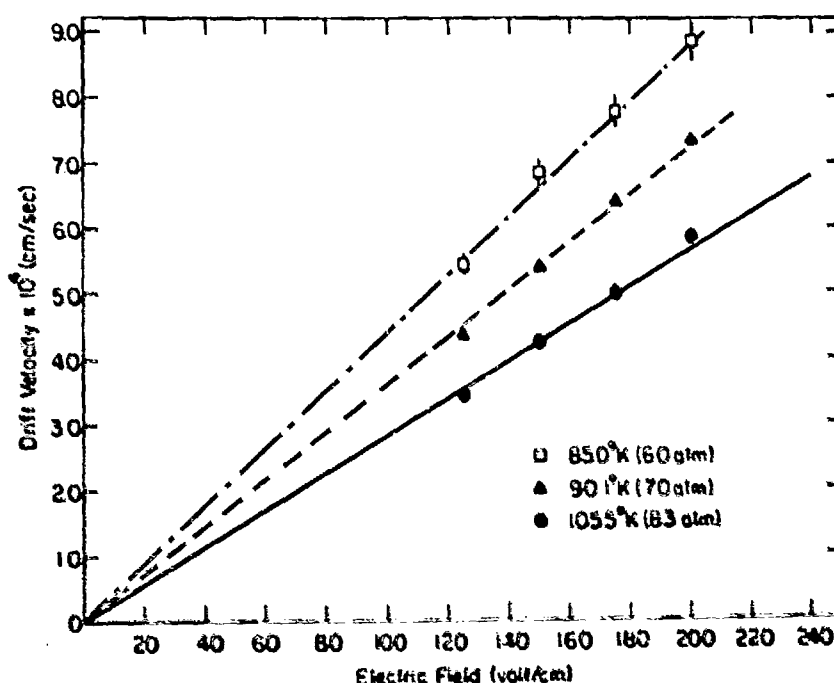
FIGURE 1. Low field drift velocity of electrons in liquid Ar.

clearly show how the mobility of an excess electron in liquid Ar is decreased by increasing temperature (for T < 115°K) and increased by increasing pressure. Also, for T < 115°K, the drift velocity is proportional to the electric field strength.
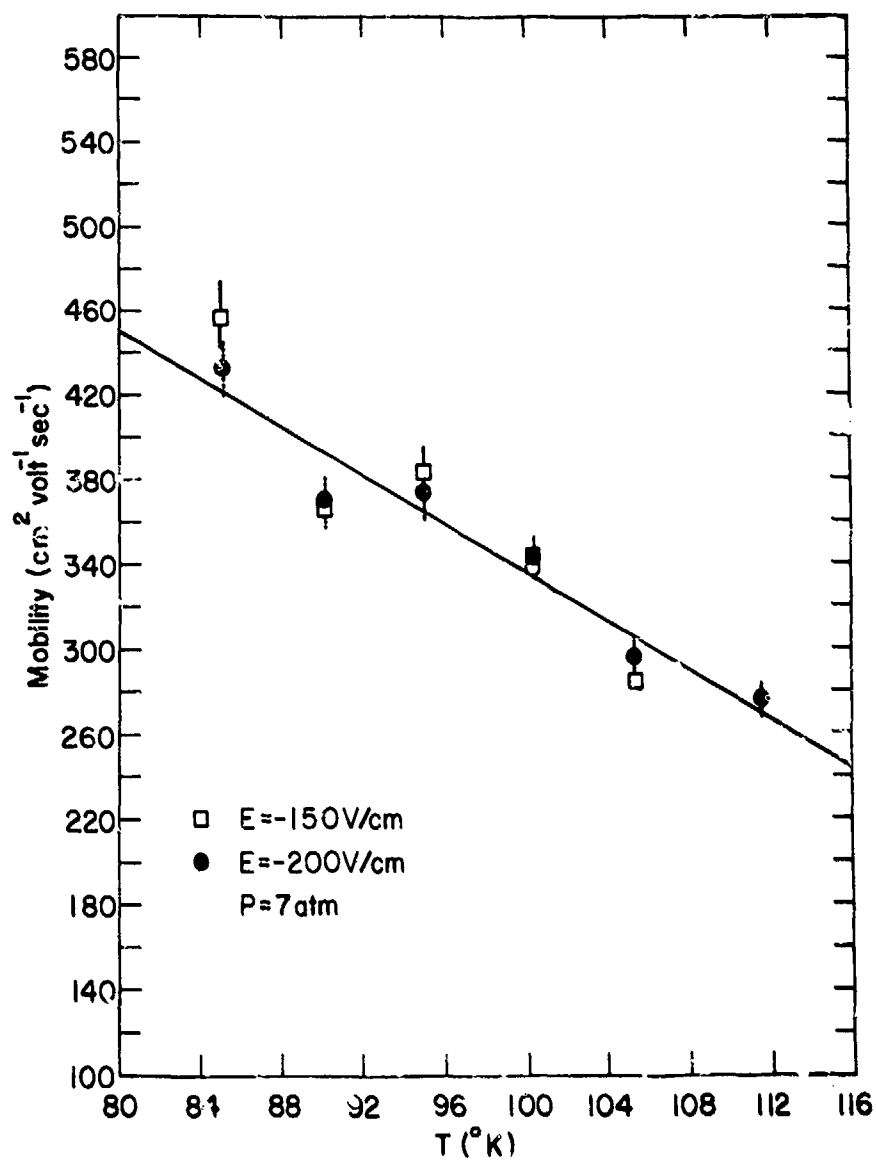
FIGURE 2. Temperature dependence of the mobility of electrons in liquid Ar.
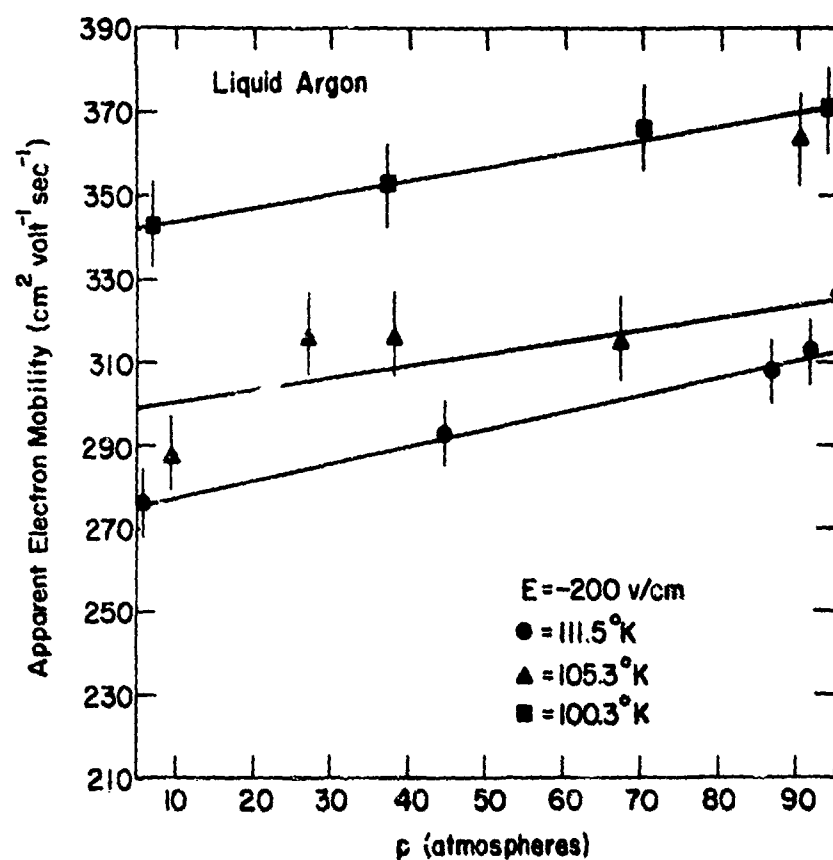
FIGURE 3. Pressure dependence of the mobility of electrons in liquid Ar.

Given the information cited, what can be said about the excess electron states of a simple liquid? We consider the development of a microscopic theory to explain these data. Clearly, there are (at least) two major problems to be solved:

(a) What is the effective atomic potential scattering the electron?

(b) Given the scattering potential, how do we determine the electron mobility and other transport properties?

An electron a distance R from an isolated atom of polarizability α induces on it a dipole of strength $\alpha e/R^2$ which, in turn, attracts the electron with a force of magnitude $2\alpha e^2/R^5$. Because of other electron-atom forces, the interaction of an atom and electron does not increase indefinitely as $R \rightarrow 0$. A convenient form for the electron-atom potential is $u_\alpha^0 = -\alpha e^2/2(R^2+R\alpha^2)^2$, where R_α is a

measure of the strength of short ranged correlation and exchange forces. The value of $R\alpha$ may be determined from the electron-atom scattering cross section in the limit of zero incident energy of the electron (scattering length). Lekner[4] has shown that when $R\alpha$ is fixed in this fashion, the electron-atom momentum transfer cross section in gaseous Ar is very accurately reproduced up to electron energies of several volts.

What is the polarization interaction between an electron and a particular atom in a liquid? To answer this question we must find the local field acting on the atom, which consists of the direct field and the sum of all other fields arising from dipoles induced on neighboring atoms. The problem is simplified by the fact that $v_A \ll v_e \ll v_{Ae}$, where v_A, v_e and v_{Ae} are typical velocities of atoms in the liquid, of excess electrons, and of the bound atomic electrons. Because of the large differences between these velocities, the motion of the atoms can be ignored in calculating the mutual screening effect of neighbors, and the motion of the excess electrons may be ignored in calculating the induced polarizations of the atomic electrons.

Consider, now, a point charge, $-e$, in a liquid composed of atoms of polarizability α. In the absence of other nearby atoms the electric field acting on an atom at R would be e/R^2 (see Fig. 4). We define
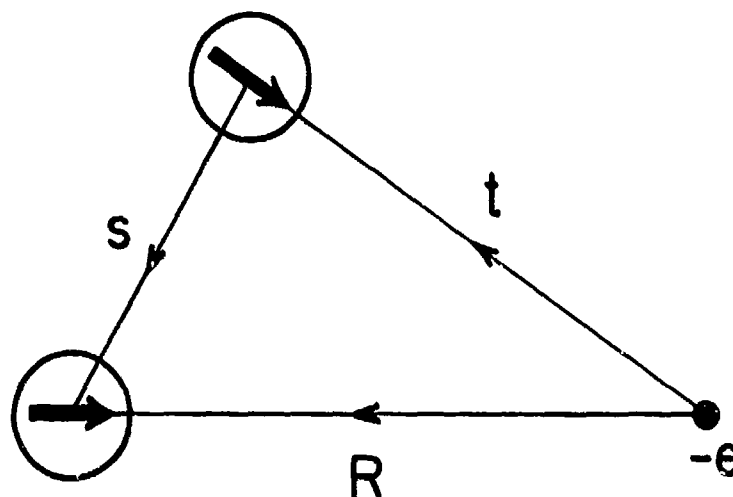
FIGURE 4. Relationships between the dipoles induced by a point charge in a simple liquid.

the average local field acting on the atom at $\mathbf{R}$, and along $\mathbf{R}$, by (e/R^2) f (R). But this local field is equal to the direct field (e/R^2), plus the contribution to the field arising from all the other induced dipoles in the liquid. Given that an atom is located at some point in the liquid, the probability of finding another atom in the volume element dt at distance s from the first is defined to be ρ g (s) dt, where ρ is the number density of atoms and g(s) is the pair correlation function (radial distribution function). The field acting on this second atom is (e/t^2) f (t), so that it carries an induced dipole of average strength α (e/t^2) f (t). After calculation of the component along $\mathbf{R}$ of the field at $\mathbf{R}$ arising from this dipole, and integration over all possible positions of the second atom, it is found that f(t) must satisfy a linear integral equation.[4] The solution of that linear integral equation gives the required self-consistent ensemble-averaged local field. The local field for a realistic liquid structure is shown in Fig. (5).[4] The variation of shielding with distance is particularly noteworthy. It is easy to show that the screening effects are contained entirely in the local field and therefore that the electron-atom polarization interaction in the liquid is $u\alpha = -\alpha e^2 f^{(\mathbf{R})} / (R^2 + R_\alpha^2)^2$. Of course, the total electron-atomic potential is a sum of the polarization and atomic potentials. For a liquid like Ar the atomic potential is little influenced by the state of aggregation, and we may assume that $R\alpha$ is the same in the gas and the liquid.

Because of the overlapping of potential fields in the liquid, the electron is never in field-free space. Now, the average potential near an atom at $\mathbf{R_1}$ is the sum of the atomic field centered at $\mathbf{R_1}$ and the average over the positions of all other atoms. The effective potential for the electron scattering is then defined by the difference between the instantaneous potential and the average potential. As shown in Fig. 6, $u_{o/f}$ is very much weaker and very much more slowly varying, than is the scattering potential in the gas phase. With the calculation of $u_{o/f}$, we have answered question (a) to the accuracy required for our present purposes.

To answer question (b) we adopt the single scattering approximation, that is, the scattered amplitude of the electron at any point is the coherent sum of amplitudes scattered from individual atoms, with neglect of the sum of amplitudes multiply scattered from different atoms. The wave incident on each atom is a wave packet which is approximated by a plane wave. The single scattering ap-
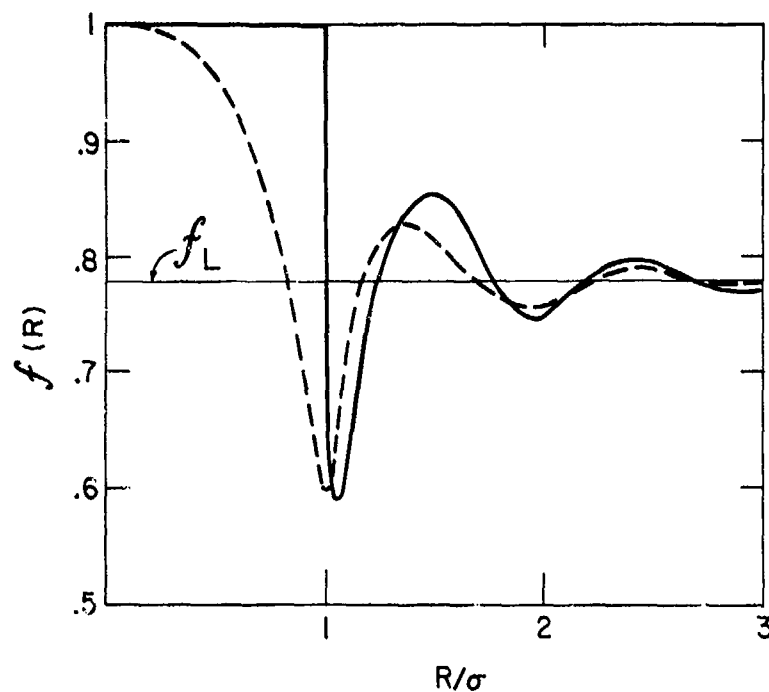
FIGURE 5. (a) The shielding function f(R) in liquid Ar.
The dashed line refers to the interaction of a point charge with neutral atoms, the solid line to the interaction of a charge with diameter equal to the atomic diameter.

proximation is valid when the mean free path, Λ, is large compared with the de Broglie wavelength of the wave packet.

As a result of scattering, a particle-wave moving through a liquid transfers energy to all collective excitations of the system. In the single scattering approximation all the necessary information about the excitations is contained in the function that describes the probability of finding a molecule at R' at time t', given that a selected molecule was at R at time t. This function is called the Van Hove space-time pair correlation function, and its Fourier transform, denoted $S(k, \omega)$, is called the spectral function.[5] The probability of an electron scattering with loss of momentum $\hbar k$ and energy $\hbar\omega$, say by creating a density fluctuation of momentum $\hbar k$ and energy $\hbar\omega$, is proportional to the product of the single atom-electron differential scattering cross section and the spectral function. Now, the spectral function has certain general properties which are
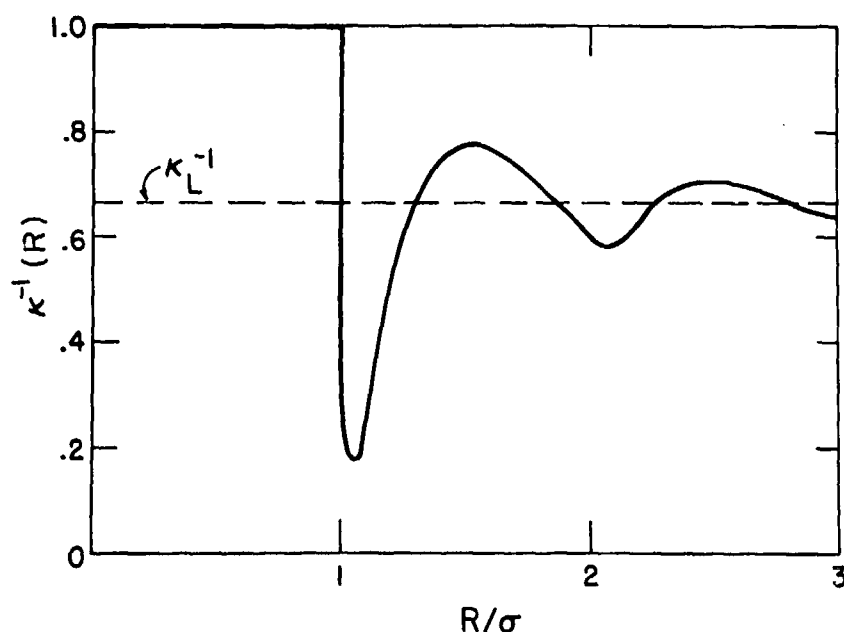
FIGURE 5. (b) The effective dielectric constant as a function of charge-charge separation in liquid Ar.

of use to us. First, the probability of scattering with momentum transfer $\hbar k$, averaged over all possible energy transfers, is proportional to the structure factor $S(k)$, which is itself just the Fourier transform of the static excess pair correlation function $G(R, o)$ $[G(R, o) \equiv g(R) - 1]$. This is, then, just the same result as embodied in the familiar formula for the intensity of X-ray scattering from a liquid.[6] Second, the average energy transfer in an interaction is exactly equal to the free atom recoil energy for the same momentum transfer, independently of structure or thermal motion. Third, it may be shown that the mean square energy transfer is only approximately structure independent. The mean square energy transfer is structure independent if we neglect, relative to the mean energy transfer, terms which are smaller by the ratio of the energy transferred in a collision to the thermal energy of an atc.. $\sim\epsilon/Mk_BT$, and hence negligible for our case.

The conditions cited can now be used to derive a kinetic equation descriptive of the electron-liquid system. The important idea is that because the electron atom mass ratio, m/M, is so small, an electron
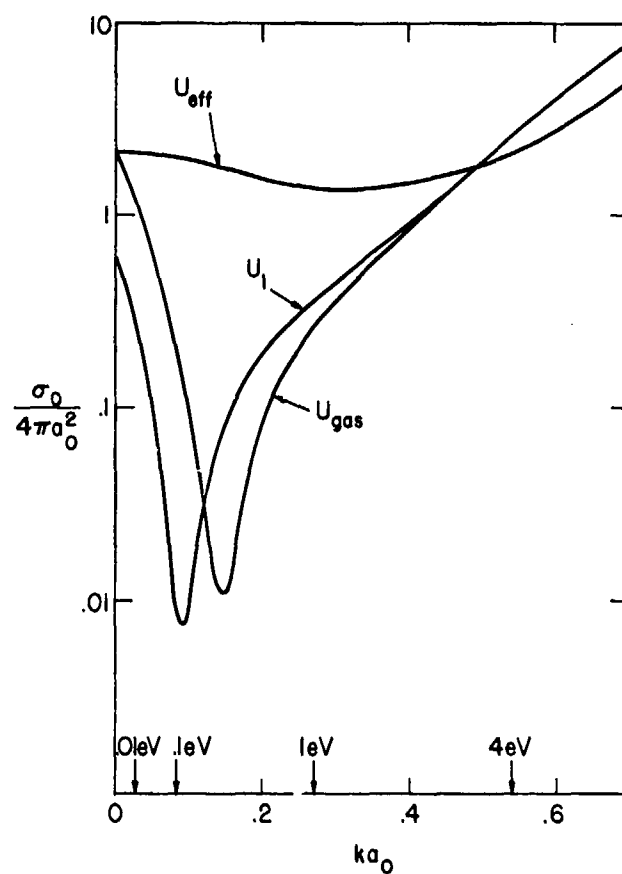
FIGURE 6. The effective potential acting on an electron in liquid Ar.

colliding elastically with an atom undergoes large deflections, but suffers only very small changes in energy. It is found that the rate of transfer of energy is determined by a mean free path

$$\Lambda_\epsilon^{-1} = 2\pi\rho \int_0^\pi \sin\theta \; (1 - \cos\theta) \; \sigma \; (\epsilon,\theta) \; d\theta \tag{1}$$

which is independent of structure, while the rate of transfer of momentum is determined by a mean free path

$$\Lambda_p^{-1} = 2\pi\rho \int_0^\pi \sin\theta \; (1 - \cos\theta) \; \sigma \; (\epsilon,\theta) \; S(\mathbf{k}_0) \, d\theta \tag{2}$$

which does depend on the liquid structure. For thermal electrons, for which $k_0 \approx 0$, the efficiency of energy transfer is greater than that of momentum transfer by the factor $1/S(0)$, which is very large in a liquid. Thus, an electron in a liquid comes to thermal equilibrium very fast, and hot electron effects require large applied fields.

The predicted drift velocity, as a function of field strength, is displayed in Fig. 7. Good agreement between theory and experiment is obtained.[4]

A different test of the theory is provided by measuring the work required to inject an electron into the liquid. The difference in energy resulting from the transfer of an electron from the vacuum to a liquid is just the mean potential acting on the electron in the liquid. By measuring the work function for emission of electrons into vacuum and into a liquid, Lekner, Halpern, Gomer, and Rice[7] find (see Fig. 8) that the energy in the liquid is −0.33 eV relative to the vacuum. The theoretical calculations sketched lead to the prediction that this energy change is −0.46 eV.

The preceding considerations suggest that a conduction electron in liquid Ar is nearly free, and that the scattering can be described in terms of the combined effects of the collective polarization field and the superposed atomic fields, at least in the first approximation. As we shall see, a necessary condition for the validity of this deduction is that the electron-atom scattering length be sufficiently small that geometric reorganization in the liquid is not energetically favored.

b. Bound Excess Electron States

When the properties of an excess electron in liquid He are studied,[8] it is found that the electron mobility is very much less than anticipated, i.e., of the order of 10^{-2} cm^2 sec^{-1} volt^{-1}. Furthermore, a study of the electron mobility as a function of density reveals that there is a drastic change as the density is increased towards the liquid density (see Fig. 9).[9] Examination of the electron-atom interaction reveals,[10] in this case, a very strong repulsion (see Fig. 10), which in turn suggests that the quasi-free electron configuration may be of higher free energy than other configurations. What other configuration might be more stable than the free electron state? An obvious possibility is that the electron-atom repulsion is strong
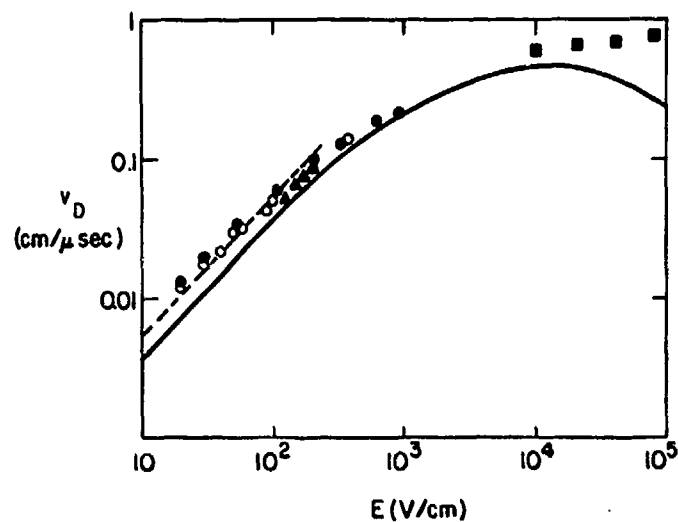
FIGURE 7. Drift velocity of electrons in liquid Ar as a function of the applied field strength. The solid curve is the prediction of the theory described in the text.
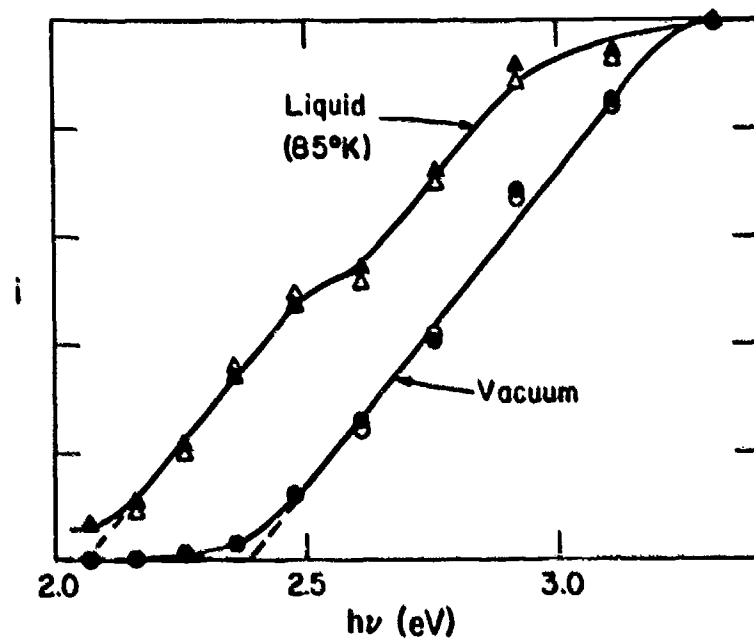

FIGURE 8. Photo-injected electron current in liquid Ar as a function of the illuminating photon energy. The surface from which the electrons come is Ba-Cs.
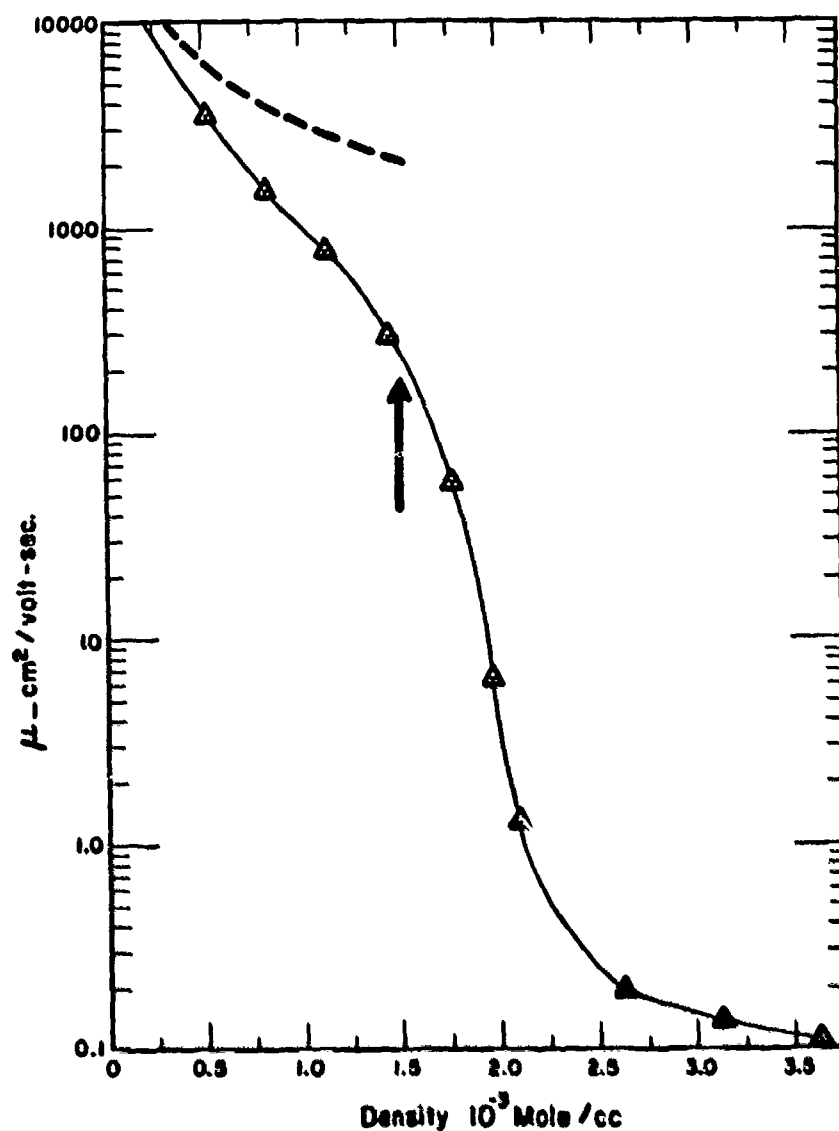
FIGURE 9. The dependence of the electron mobility on gas density in He.

energy to create a void around the center of the electron wave pack-
et. The work required to create this bubble depends on the volume
of the bubble, the effective surface tension of the liquid (new sur-
face area is created when a bubble is formed) and the increase in
electron kinetic energy because of localization inside the bubble.
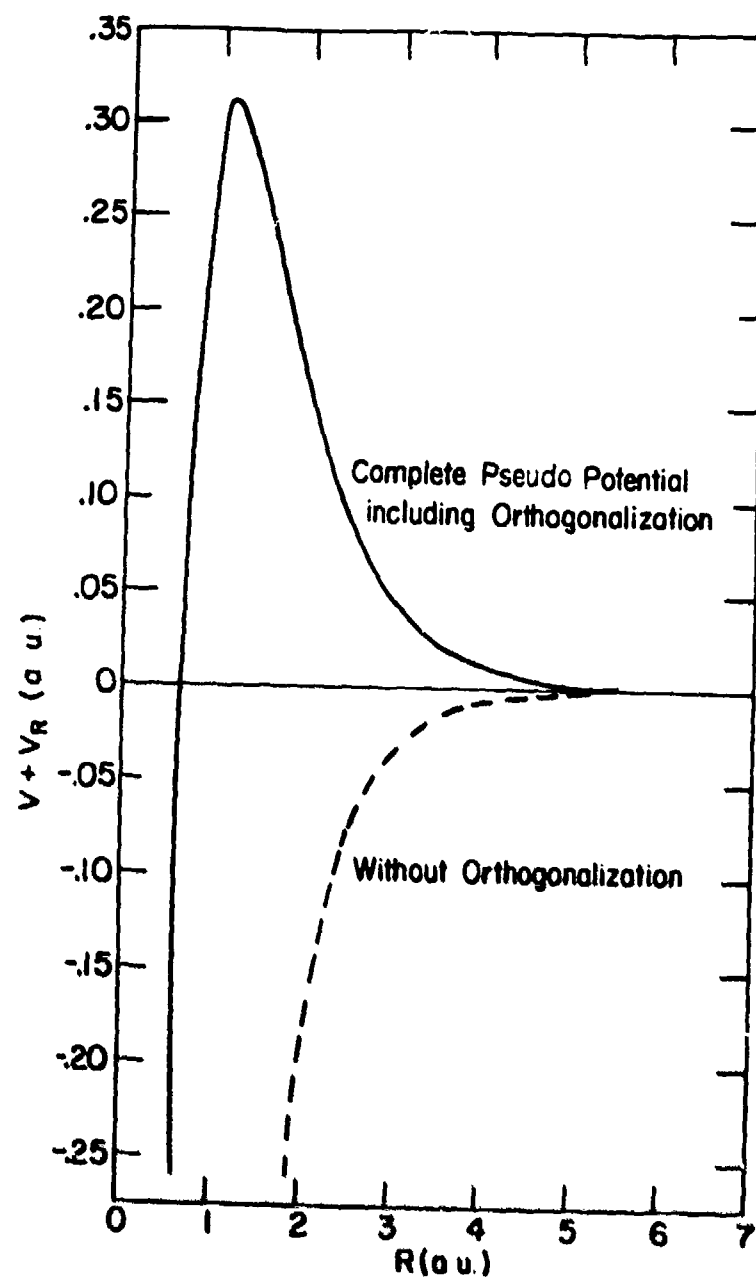
Figure 10. The effective electron-He atom repulsion.

Of course, the electron-atom repulsion is reduced as a result of enclosing the electron in a void, and whether or not bubble formation is thermodynamically favored depends on the balance between all the factors mentioned. The bubble model was first suggested by Careri and Feynman[11] and has since been studied by several other investigators. The most sophisticated calculations are those of Hiroike, Kestner, Rice, and Jortner,[12] who use the mathematical isomorphism between a pair product form of the wave function and the pair distribution function of a classical liquid in an external field to avoid the introduction of a surface tension, etc. All calculations are in agreement that:

(a) Void formation is only favored at high density (see Fig. 11).
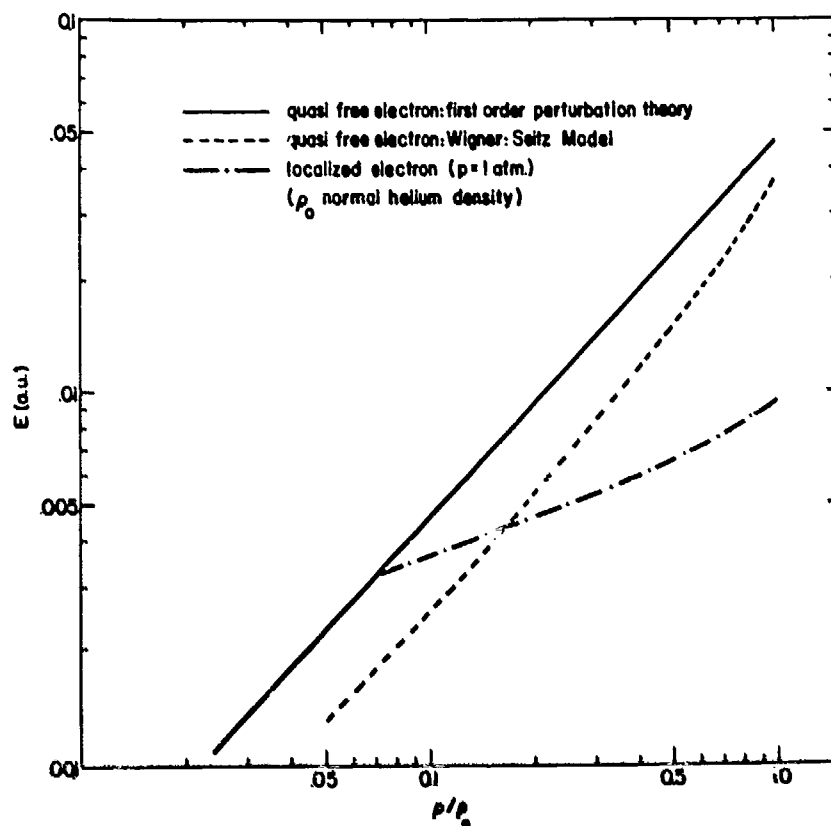
FIGURE 11. The free energies of a free electron and a bound (bubble) electron in He as a function of density. Note that the density at which the curves cross is close to the density marked by an arrow in Fig. 9.

(b) The qualitative features of the trapped state are independent of whether or not the bubble boundary is fuzzy or sharp (see Fig. 12) and of the gross magnitude of the surface energy (see Fig. 13).
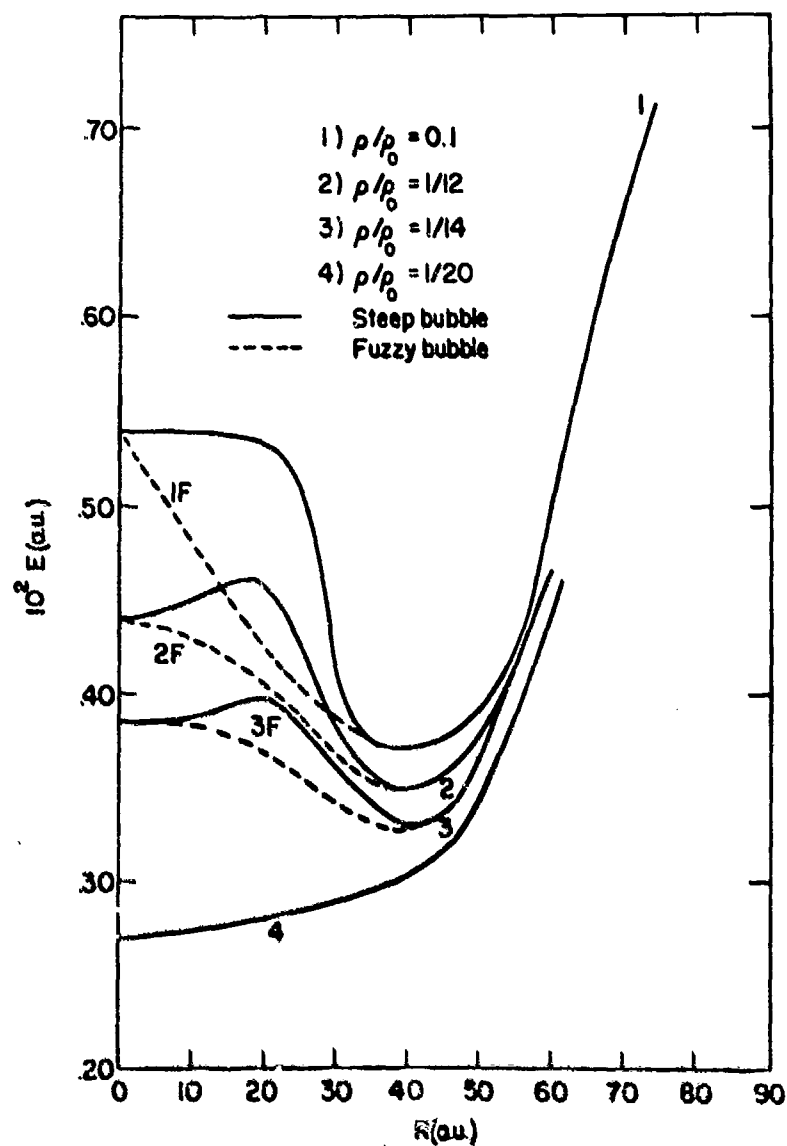
Figure 12. The effect of altering the bubble boundary density (fuzzy boundary) on the stability of the bubble configuration in He.
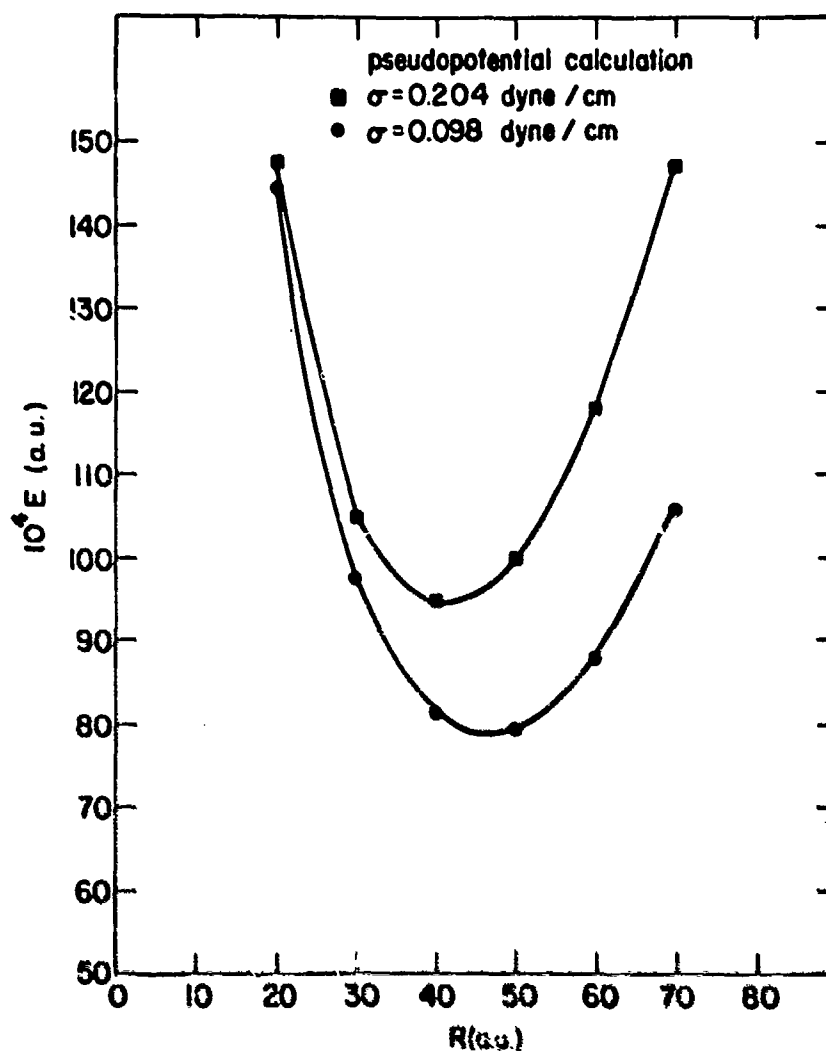
Figure 13. The effect of surface tension on the stability of the bubble configuration in liquid He.

In the calculations of Jortner, Rice, Kestner, Hiroike, and Cohen[12] the electron-atom interaction was calculated from pseudopotential theory. In quantitative terms, these calculations account for:

(c) the mobility of the electron and its pressure and temperature dependence.[13]

(d) The energy required to inject an electron into liquid He (theory 1.0 eV, observed 1.1 eV).[14]

(e) The density at which the transition from delocalized to localized electron states occurs (see Fig. 9),

(f) The size of the void as measured by electron-vortex line trapping experiments.[15]

Thus, in contrast to the case of liquid Ar, in the case of liquid He the excess electron states are nothing at all like free electron plane wave states. We see, then, that the excited states of a simple liquid are complex and depend very strongly on the nature of the electron-atom interaction. It is just because the lack of rigid lattice permits easy geometric readjustment of the local structure that the electronic states are so strongly coupled to the translational states of the liquid. We may anticipate a rich variety of behavior of excess electron (conduction electron) states, dependent on details of the electron-molecule interaction and perhaps on the nature of the internal states of the host molecules. We are only at the very threshold of understanding what these states are like or how they behave under external perturbation.

Thus far in Sections 2a and 2b we have discussed those properties of the conduction electron states which are, at least qualitatively, understood. Examples of phenomena that are not understood include the following:

(a) In liquid Ar above $T = 115°K$ and in liquid Kr for all temperatures thus far studied, the temperature dependence of the drift velocity is opposite to that expected (see Figs. 14 and 15).[3] If it is a valid argument that the polarization field in the liquid sums to an almost constant field which does not influence the scattering, one cannot invoke a Ramsauer effect to explain these data. The fact that the drift velocity depends on the field strength more than linearly suggests that some feature of the scattering process is not properly accounted for in the analysis thus far developed.

(b) There is observed to be a transition from localized excess electron states to delocalized excess electron states in systems such as Na in molten NaCl.[16] The nature of this transition is intimately related to the interplay between electron-electron and electron-ion (or atom) interactions. Many theoretical studies of the non-metal-metal transition have been made,[17] and there is relevant experimental data from studies of impurity conduction in doped semiconductors.[18] However, these studies do not describe the role of the local ion or atom structure, and more particularly the possibility of
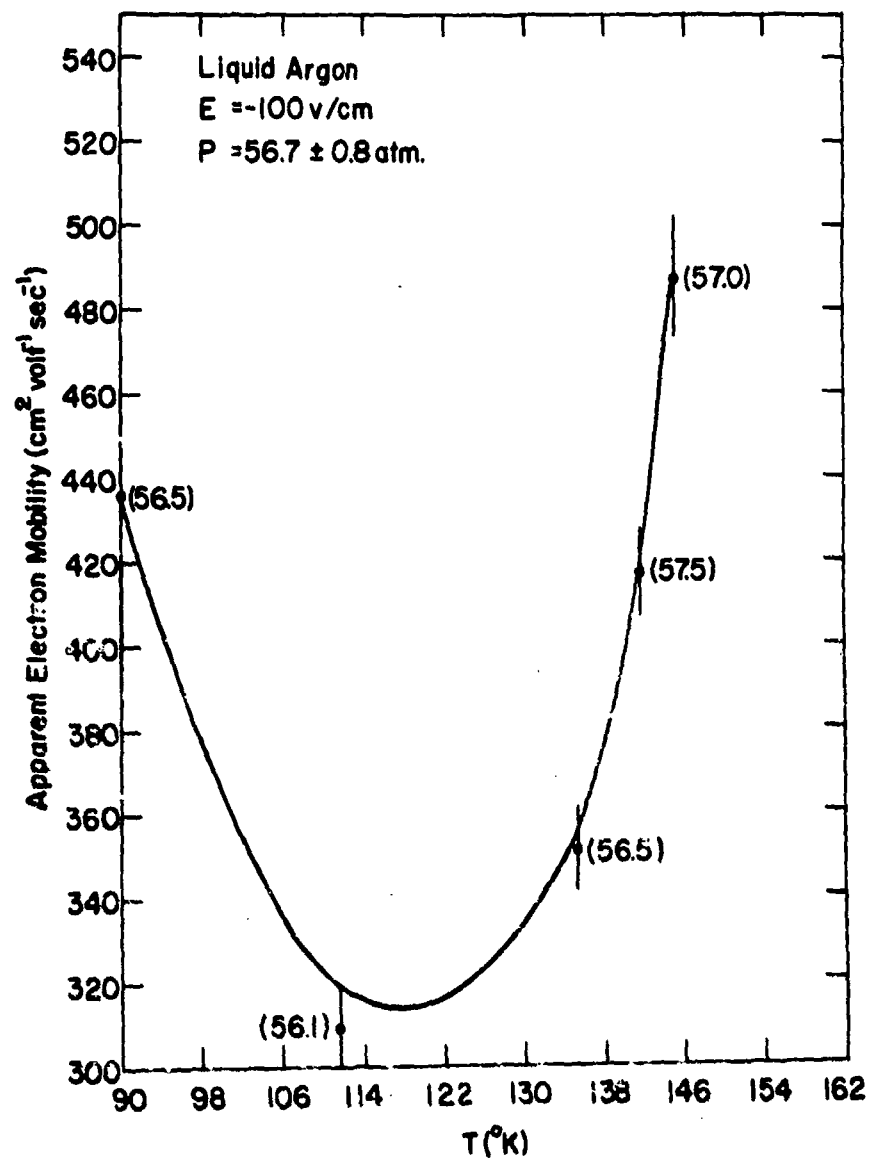
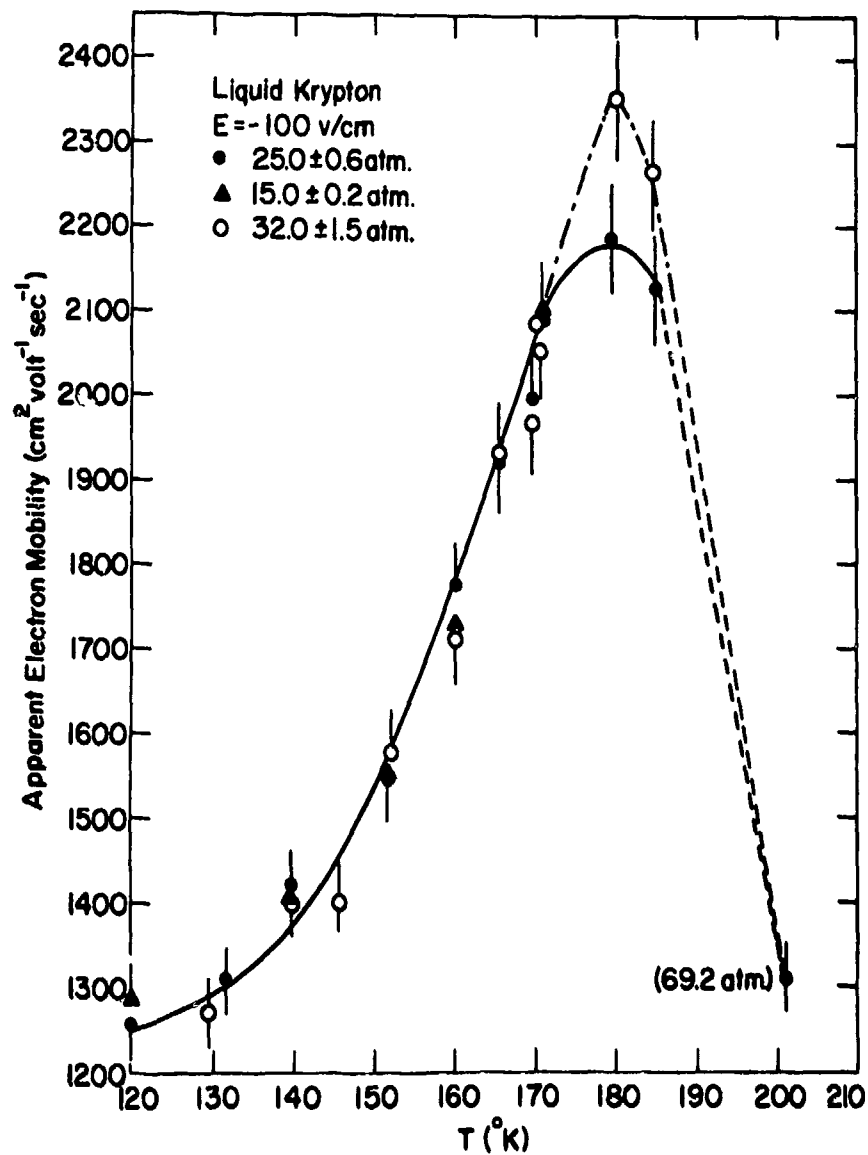FIGURE 14. The effective mobility as a function of temperature in liquid Ar.

FIGURE 15. The effective mobility as a function of temperature in liquid Kr.

local deformation, in determining the transition, since all use a lattice model with an assumed random distribution of impurity centers. It is possible that the differences between the electronic struc-

tures of the doped solid and liquid are as great as those between a pure solid and liquid. There are also studies of the properties of liquid metals,[17] but these use approximations not necessarily valid in dielectric liquids.

The solution of these and other problems may require new constructs and lead to new and more incisive understanding of the conduction electron states of all media. The problems which will be raised when systems with internal states, e.g., polyatomic molecules, polymers, etc., can now only be foreseen with difficulty, but they too should be of great intrinsic interest and value in the construction of a complete theory of the electronic properties of matter.

3. EXCITON STATES IN A MONOATOMIC DIELECTRIC LIQUID

We now examine some of the properties of the bound excited states of a simple liquid. It is convenient to consider several interrelated questions:

(a) Do exciton states exist in a liquid? If such states do exist, what is the nature of the spectrum?

(b) How do intermolecular interactions alter the spectrum of stationary states?

(c) How is energy transferred in a liquid?

(d) What is the nature of the relationship between the bound electron states and free electron states in the liquid?

Clearly, questions (a) and (b) are intimately coupled to one another. Nevertheless, it is useful to proceed by first considering the properties of the bound states in the absence of scattering, and then to examine how the spectrum of states is altered by scattering processes.

a. STATIONARY STATES IN THE ABSENCE OF SCATTERING

From the most general point of view, it may be argued that on the scale of length determined by the wavelength of typical electronic transitions, both liquids and solids display translational symmetry. Indeed, it is only for distances of the order of 5-50 A that differences in the local geometries of liquids and solids are obvious. Thus, provided that the wavelength of an incident electromagnetic wave is large relative to the range of molecular ordering, localized excitations at two points in the liquid are related by the phase factor exp

(i k·R), where **k** is the excitation propagation vector and **R** the vector separation of the two points. We shall see later that even in the absence of scattering, the disorder in the medium leads to damping, but this can be shown to be small in the limit that $|k|$ is very small relative to the reciprocal of the near neighbor distance.

To describe the internal structure of a possible exciton state we utilize the deductions of Section 2a; that is, we assume that an electron is nearly free in Ar and, therefore, that the wave function of a conduction electron in liquid Ar is adequately represented by a plane wave. It is also convenient, but not necessary, to assume that the hole (ion core) is stationary. Although the hole will have finite mobility, in general it moves much more slowly than does the electron. Now the simplest approximation to the dynamics of the hole-electron pair is the following. A wave packet describing the electron is constructed by superposing, with appropriate coefficients, the free electron plane wave eigen-functions. The Hamiltonian of the electron hole pair is then represented as a sum of the free electron Hamiltonian and the screened coulomb interaction. It is then readily shown that[19]

$$\frac{\hbar^2 k^2}{2m} A(k) + \sum_{k'} \tilde{u}_{k \cdot k'} A(k') = EA(k) \qquad (3)$$

where $A(k)$ is the coefficient of the plane wave $|k>$ in the electron wave packet, and $\tilde{u}_{k \cdot k'}$ is the Fourier transform of the screened coulomb potential. If u were a simple coulomb potential, (14) would be the momentum representation of the hydrogenic Schroedinger equation for the amplitudes $A(k)$. In this limit, the manifold of levels is hydrogenic, and the amplitudes in the wave packet expansion satisfy a hydrogenic equation. The results of Section 2a clearly show that u is not a coulomb potential, and therefore the energy level structure deviates from the hydrogenic structure. Using the screened coulomb interaction appropriate to liquid Ar, obtained by methods similar to those described earlier, the eigenstates of Eq. (14) having s symmetry have been determined.[29] Some calculated charge densities are shown in Fig. 16 together with the corresponding hydrogenic charge densities. Clearly, the shifts in charge density are just those to be expected from the modified form of the coulomb interaction.
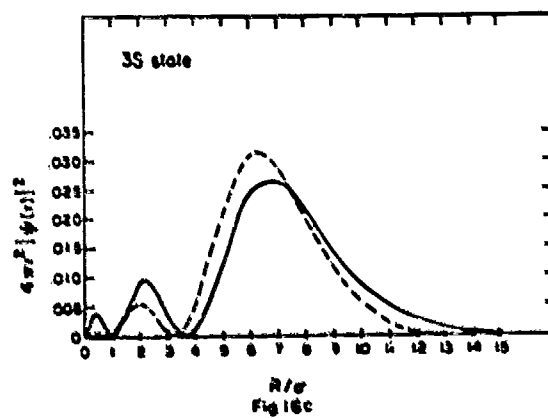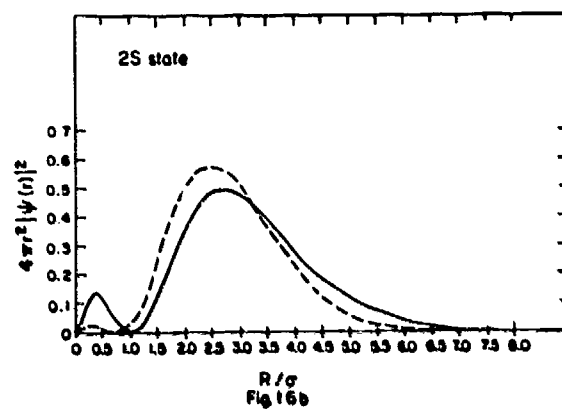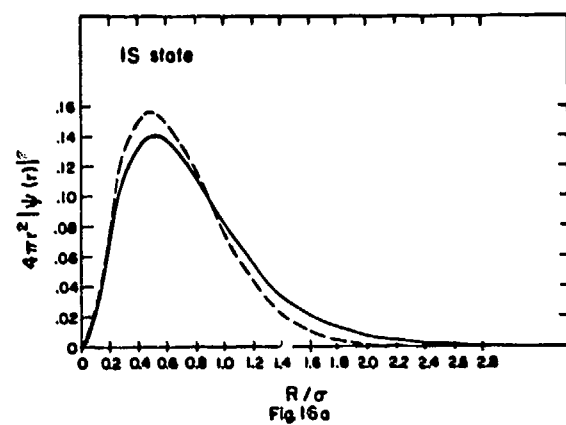
FIGURE 16. The charge density in s states of a quasi-hydrogen center in liquid Ar.

The preceeding simple calculations in no way elucidate the dependence of the energy of the exciton on the propagation vector, called the dispersion relation. To study the dispersion relation we resort to a different set of considerations. Suppose some one molecule in the liquid is excited, and that there is a nonvanishing dipole matrix element between the excited state and the ground state of the molecule. Then, because all the molecules in the liquid are identical, and each of the excited states of the N molecules is N-fold degenerate, excitation energy may be transferred from molecule to molecule. This resonance transfer of energy may be described in terms of the coupling between the transition dipole moments of the molecules. Because of the long range of the dipole interaction, dipolar motion is organized into collective dipolar polarization waves. In turn, the polarization waves may be interpreted as the classical equivalent of an exciton field. It may be shown, for example, that the van der Waals energy of a medium of nonoverlapping molecules arises from the shift in the spectrum of dipolar fluctuations under the influence of their mutual interactions.[21] In making this last statement we have emphasized that the molecules concerned must be such that there is vanishingly small overlap of the electronic wave functions in both the ground state and the excited states. If there is overlap between the molecules when one is excited, additional interactions not included (e.g., charge transfer) must be of some importance in determining the cohesive energy and other properties of the system. In the following we confine attention to the case that overlap of electronic wave functions is vanishingly small.

To compute the exciton dispersion curve in a simple liquid Nicolis and Rice[22] make use of the analogy between the classical polarization field and an exciton field. They consider an assembly of N atoms (molecules) in a volume V, and represent each atom by a Drude atom, i.e., each atom is assumed to have s bound electrons of charge $e f^{1/2}_n$ ($n = 1, 2, \ldots, s$). Furthermore, each of the electrons is assumed to undergo undamped harmonic oscillation about the nucleus with frequencies ω_n ($n = 1, 2, \ldots, s$). The connection between this classical model and the correct quantum mechanical description is established by requiring f_n to be the oscillator strength corresponding to the transition of interest. To complete the specification of the model system we must describe the nature of the in-

teratomic interactions. Nicolis and Rice assume that the total potential energy of interaction consists of:

(a) A dipole-dipole interaction between the instantaneous transition dipoles on each atom,

(b) A superposition of pair interactions corresponding to short range repulsions and whatever residual high order multipolar interactions may exist in the system.

The system just described has two features of interest. First, the equations of motion for the amplitudes of individual atomic dipoles, including the coupling terms, are linear when represented in the complete phase space of the system. Second, the range of the transition dipole-transition dipole interaction is very large relative to the range of the other interactions between atoms, and this interaction is also weak relative to the strength of the other interactions. The first observation suggests that the dipole amplitudes oscillate harmonically. The second observation enables Nicolis and Rice to calculate the dispersion relation for the system in terms of an expansion in which the parameter is the ratio of the ranges of the short and long range interactions. This parameter is very small in the case considered. It is found that the spectrum has transverse and longitudinal modes and that the dispersion relation has the following interesting properties:

(a) The two transverse branches of the dispersion relation differ only through the difference between the unperturbed modes, ω_{at}' ω_{at}''.

(b) When the wave vector k is small, the change in spectrum introduced by the dipole-dipole coupling is parabolic in k.

(c) There is a gap in the spectrum of transverse polarization waves at $k = 0$, with the magnitude of the gap dependent on the magnitude of the dipole-dipole coupling.

(d) In the limit $\frac{\pi}{2} < |k| \sigma < \pi$ (σ is a molecular diameter), there is a change in sign of the frequency shift arising from dipole-dipole coupling and the frequency shifts of the longitudinal and transverse modes are of opposite sign. The change in sign results from a reversal of the orientation of the dipoles which are near neighbors to any selected dipole when $|k|$ passes into the indicated range from larger to smaller values of $|k|$.

(e) An examination of the imaginary part of the dielectric function shows that the longitudinal polarization waves are damped,
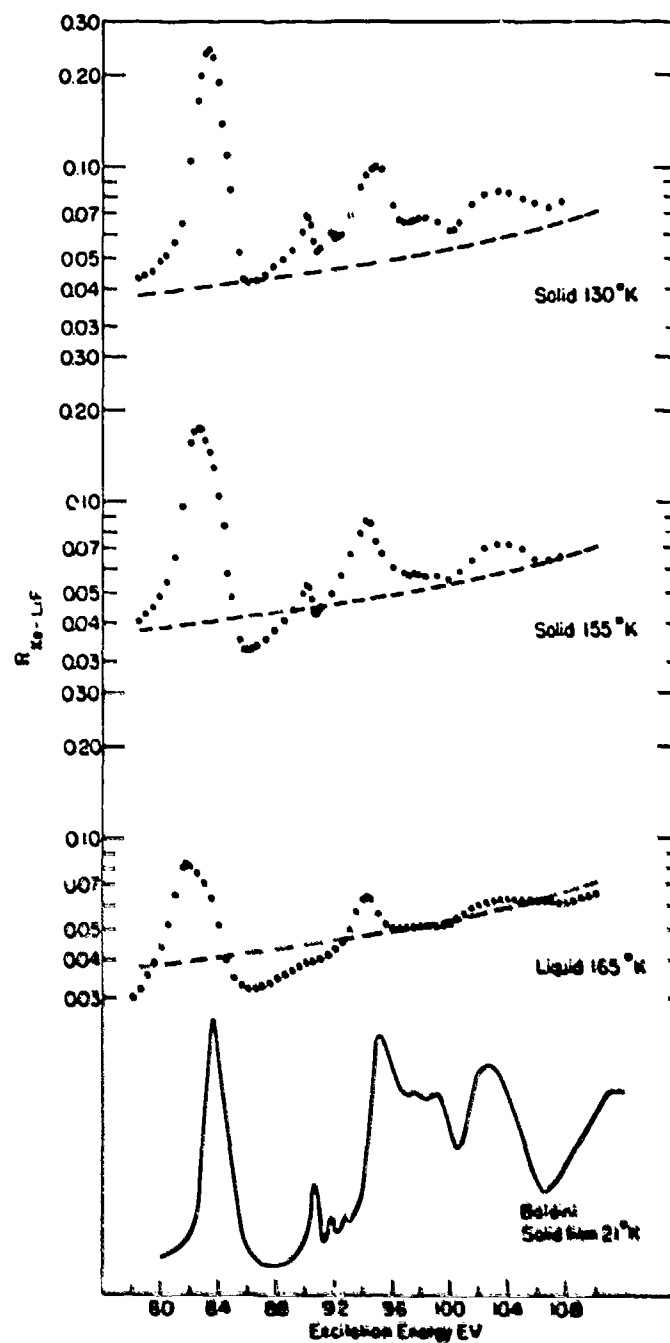
Figure 17. The reflection spectrum of liquid Xe.

but that in the limit $\mathbf{k} \to 0$ the damping is small. The damping arises from phase mixing which leads to instability of the collective polarization wave relative to the independent dipole oscillations, and is the analogue of Landau damping in a plasma.[23] Thus, in the absence of real scattering, we are justified in considering the polarization waves to be quasi-normal modes when $\mathbf{k} \to 0$, but because the Landau damping increases greatly as $|\mathbf{k}| \to \sigma^{-1}$ this interpretation would not be valid as $|\mathbf{k}| \to \sigma^{-1}$. Since optical excitation in the visible and ultraviolet regions corresponds to $|\mathbf{k}|\sigma \ll 1$, we conclude that in the absence of real scattering events in the liquid, excitons can exist despite the disorder in the liquid phase.

The limitations and approximations of the theory of Nicolis and Rice resulting from the simple model used are, with one exception, of secondary importance. The one exception is the neglect of all scattering events. Although it has been shown that in the long wavelength limit there exists a damping of the polarization waves arising from phase mixing, it is obvious that there must exist other, more efficient damping mechanisms. For the case of electronic excitation these will arise from the scattering of the excited electron by the fields of the surrounding atoms, and are analogous to electron-phonon coupling in the crystalline solid. We, therefore, expect there to be a shift and broadening of the exciton spectrum calculated for the case of no scattering. Before examining the limited experimental data we consider this problem.

b. The Shift and Broadening of the Excitation Spectrum

The simplest description of the effects of scattering is in terms of the hydrogenic model discussed at the beginning of Section 3a. The argument leading to the conclusion that bound states exist is of considerable generality, relying only on the existence of plane wave states for the free electron. The bound states of the core-electron pair will be reasonably well defined if the mean free path of the electron is significantly larger than the orbital circumference. Scattering of the orbital electron by the atoms of the liquid causes a decrease in the lifetime of any given state and, in the limit that the scattering is so frequent that an orbit cannot be closed, no bound state can exist. This limiting case is, of course, inconsistent with the assumption that the free electron is in a plane wave state.

Let us assume that the scattering of the electron is sufficiently weak

that bound states do exist. We propose to compute the line width by setting[19]

$$\Delta \nu = \frac{1}{\pi \tau} \tag{4}$$

where the relaxation time, τ, includes the effects of coherence between the scattering amplitudes from different centers. Rice and Jortner[19] assume (4) to be valid, and use for τ the relaxation time for momentum transfer considered in Section 2a. When the scattering potential is represented in terms of the zero energy scattering length, a, they find

$$\frac{1}{\tau (k)} = \frac{\rho \hbar^2 a^2 k}{m} S(k) \tag{5}$$

Because the magnitude of the electron wave vector in a bound state is related to the orbital velocity by $|k| = m |v| / \hbar$, we conclude that the scattering time is inversely proportional to the speed of the electron. Thus, the electron makes fewer collisions in the tightly bound states, and the lifetimes of the states should decrease as the principal quantum number increases.

An immediate test of (4) and (5) is possible using the reflection spectrum of liquid Xe measured by Beaglehole.[24] From the available electron-atom cross sections we are led to the prediction that hydrogen-like levels will be broadened, in liquid Xe, by about 0.1 eV. Since the spacing between levels is only about 0.2 eV,[25] the set of exciton levels will appear as an unresolved spectrum. As seen in Fig. 17 this is in agreement with what is observed—the total width of the exciton manifold is about the same in liquid and solid, but resolution of the level structure in the liquid is not possible.

Although the preceding arguments are informative, they do not come to grips with the fundamental problems which arise in describing the influence of scattering events on the exciton spectrum. These difficulties are of two kinds:

(a) It is necessary to describe dynamical processes in a liquid, for which the simplifications provided by the binary collision approximation (valid in a dilute gas) or the simple exciton-phonon linear interaction (valid in a class of crystals) are not available.

(b) There is coupling between the resonant interactions and non-resonant interactions in the medium.

At present no fully satisfactory solution to these problems is available, but there do exist approximate calculations that suggest which physical phenomena are of importance. Popielawski and Rice[20] have studied an approximation, based on the Fano theory of line broadening,[1] to describe the shift and broadening of impurity spectra in a liquid. In their study of the impurity spectrum Popielawski and Rice assume that:

(a) The internal states of the perturbing host molecules do not influence the internal states of the guest molecule,

(b) The translational motion-internal state coupling may be represented as a sum of pair interactions,

(c) The pair interaction may be meaningfully (even if formally) separated into a strong short range component and a weak long range component,

(d) Guest-guest interactions may be neglected,

(e) The initial state of the system may be represented by the product of the density matrix for the internal state of a guest molecule and the density matrix for the translational states of all molecules in the liquid,

(f) The strong short ranged component of the intermolecular force leads to dynamical events which may be described by a modified t-matrix (binary collision) expansion,

(g) The weak long ranged component of the intermolecular force leads to dynamical events which may be described by a weak coupling expansion.

The analysis is designed to provide an approximate representation of the line shape function descriptive of transitions localized on an impurity molecule present in a simple liquid. The general physical picture which emerges from the analysis is the following: the internal states of an impurity molecule in a simple liquid are influenced by a mean field arising from the superposed long range components of the guest-host interactions of many molecules, and also by quasi-binary encounters arising from the near approach of one guest-host pair moving in the fluctuating force field of all the other molecules. In the quasi-binary encounters the important component of the interaction is short ranged. It is important to note that both the mean field effect and the quasi-binary encounters are defined so as to include the structure of the liquid in the local equilibrium approximation. The quasi-binary collision term con-

tains the influence of all sequences of successive dynamically uncorrelated encounters. A discussion of the implications of the assumptions cited, and a description of the relationship between the Popielawski-Rice description and the kinetic theory of liquids[27] may be found in reference 26.

Although the preceding arguments lead to a simple description of the relationship between the line shape function and the intermolecular interactions for the case of impurity spectra, they are not readily extended to the case where resonant interactions are important, i.e., the pure liquid. Even in the absence of overlap of the electronic wave functions of neighboring molecules, the mixing of effects arising from resonant and nonresonant interactions leads to complicated coupling phenomena. Considerable insight into the general structure of the theory can be obtained from an examination of a simple model. Though the properties of the model considered depart in detail from the properties of real systems, the most important consequences of the mixing of resonant and nonresonant interactions can be elucidated and the complications attendant to r complete (but as yet unavailable) analysis avoided.

Using a kinetic equation approach, Nicholis and Rice[28] have extended the previously described analysis of the spectrum of polarization waves in a liquid of Drude molecules. As in the Popielawski-Rice treatment,[26] the interactions between the molecules are separated into components, in this case a short-ranged repulsive interaction, a resonance interaction represented as a sum of transition-dipole-transition dipole couplings, and a residual soft (multipolar) interaction. In the case of an impurity spectrum the derived kinetic equation displays the effects of scattering from the short ranged repulsions in a modified Enskog kernel and from the residual soft potential in a Fokker-Planck-like term.[27] It is found that, even when the short ranged repulsions are represented as hard core interactions, there is both a shift and a broadening of the spectrum. The average soft field contributes only a shift in the oscillator frequency, and fluctuations about the mean field lead, again, to both a shift and broadening of the spectrum. These deductions are in agreement with those of Popielawski and Rice.

Of most interest, however, is the influence of the resonance coupling in the pure liquid, which was not treated by Popielawski and

Rice. It is found that the dipole-dipole coupling of a pair of molecules is shielded by the presence in the liquid of other molecules which are also coupled to the pair by dipolar interactions. The shielding depends on both the dipolar interactions and the short range order in the liquid, as well as the internal state of the molecules. Further, the kinetic equation contains a term which plays the role of a friction coefficient. The evolution of the internal states of a molecule is now described by an operator which includes: (a) all dynamical events corresponding to interactions in the continuum states lying above the bound states that take part in energy transfer, (b) the scattering of molecules in excited bound states, in general with excitation transfer, and (c) the formation of new bound states. The events described by (a), wherein all bound states remain unchanged and there is a modification of the continuum states arising from scattering, correspond to the limit in which the internal degrees of freedom of the molecule are organized into Frenkel-type excitation waves, i.e., although collective states of the liquid exist they have one to one parentage in the states of the free molecule. In contrast, (c) describes the formation of Wannier excitons, and (b) an intermediate case.[29] Although the theory, in principle, permits passage from the tight binding Frenkel limit to the weak binding Wannier limit, the neglect of overlap and exchange effects limits the accuracy of the description of the transition. For this reason, Nicolis and Rice confine attention to the Frenkel limit. In this limit, it is shown that the resonance interaction leads again to both a shift and broadening of the spectrum of polarization waves. The dispersion relation still displays both longitudinal and transverse branches, and a preliminary estimate of the efficiency of resonance coupling versus the internal state-translational motion coupling suggests that the friction associated with the resonance coupling is smaller than the friction associated with internal state-translational motion coupling. The theory described is too general and involves too many approximations to make any but simple deductions of the type described.

The experimental data available are inadequate to test even the limited theory described. As previously mentioned, Beaglehole has studied the reflection spectrum of liquid Xe and compared this with the spectrum of solid Xe. There is evidence for a collective

excitation in the liquid corresponding to the known Wannier-like excitations of the solid (see Fig. 17) and the inferred level broadening is about that predicted by the simple theory. More recently, Jortner and co-workers[30] have studied the absorption spectrum of Xe in Ar and have also shown that the broadening of the impurity spectrum is in agreement with the predictions of the simple theory.

Given that exciton states do exist in a liquid, i.e., that molecule scattering events do not so shorten the lifetime of collective excitations that these cease to be meaningful in the description of the liquid, it is to be expected that energy can be transferred over long distances. There exist, at present, no detailed studies of energy transfer in simple liquids. Meyer, Jortner, Rice, and Wilson[31] have studied the consequences of exciting liquid He, Ne, Ar, Kr, and Xe with α particles. The emission spectra of all the liquids are red shifted (from 2 eV to 6 eV) and, by comparison with known gas spectra, the emitting species is identified in each case as the excimer Ne_2^*, Ar_2^*, ... (see Fig. 18). The binding energy of the He_2^* is 2.6 eV,[32] and of the other excimers presumably less, but enough to promote a change in local liquid structure resulting in trapping of the excitation energy. If the lifetime of the molecular excited state is long relative to the time required for molecular displacement, and if an excimer can be formed, it seems likely that excitation energy can be self-trapped with high efficiency. Clearly, the situation in which self-trapping is generated by exciton-fluid coupling is analogous to the electron self-trapping in liquid He. Just as the excess electron states of He and Ar are fundamentally different, corresponding to trapped and free electrons, we must expect to find liquids in which energy transfer does occur with ease. Each case must be examined separately.

Of course, if the excimer species is long lived, it can serve as the carrier of energy. Indeed, a phenomenon attributable to energy transfer via intermediacy of He_2^* was discovered by Meyer, Jortner, Wilson, and Rice.[33] When liquid He was doped with N_2 and O_2 (present as small solid particles) the emission spectrum of the α-particle irradiated liquid arise from the transitions $A^3 \Sigma_u^+ \rightarrow X^1 \Sigma_g^+$ and $C^3 \Sigma_u^+ \rightarrow X^3 \Sigma_g^-$ of N_2 and O_2, respectively (See Fig. 19). These are the transitions that would be excited by triplet He_2^*, and since the lowest triplet state of He has a very long lifetime (many sec-
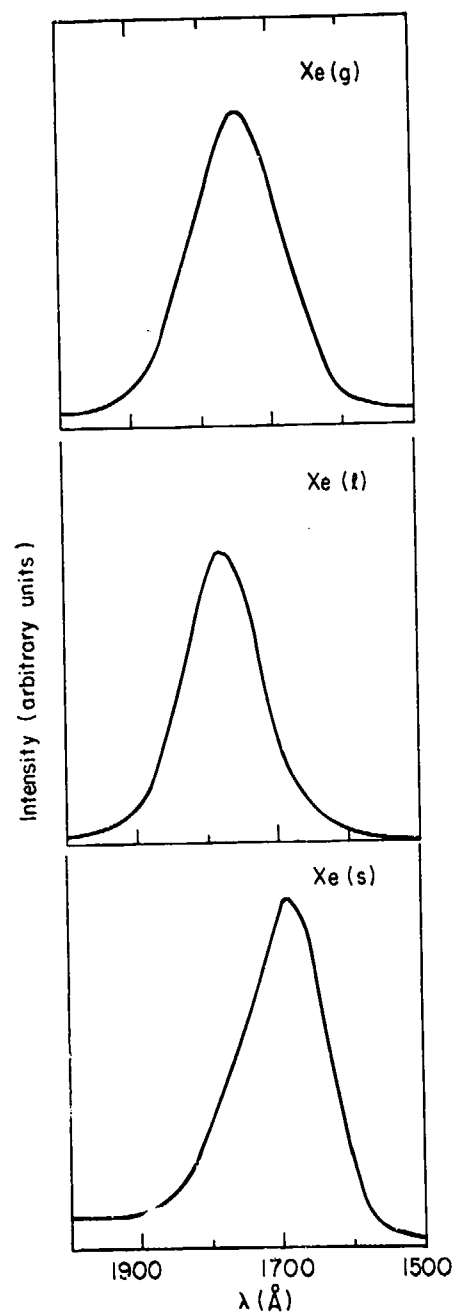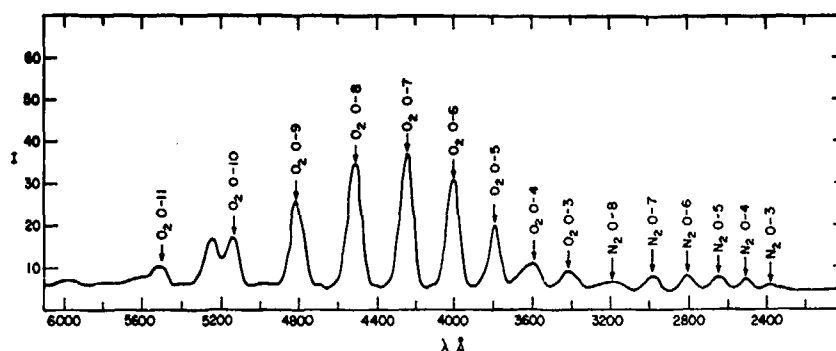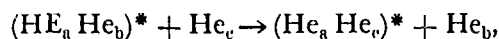
FIGURE 18. The emission spectrum of Xe.

FIGURE 19. The emission spectrum of liquid He doped with O_2 and N_2.

onds) it is reasonable to suppose that diffusion of He_2* can serve to transfer electronic excitation energy over long distances in liquid He. It should be noted that we cannot, at present, rule out atom interchange-energy exchange,[3]

$$(HE_a He_b)^* + He_c \rightarrow (He_a He_c)^* + He_b,$$

or other mechanisms of energy transfer. Much more experimental work will be required before energy transfer in liquids can even be outlined for detailed study, let alone interpreted completely.

It is clear that there remain many major problems in the description of the exciton states of a liquid. These include:

(a) Development of a formalism that allows the treatment of overlap and charge transfer phenomena,

(b) Development of an understanding of the relationship between scattering processes and excimer formation,

(c) Accumulation of a body of experimental data with which to test the ideas thus far put forward and to guide the development of an improved theory,

(d) Development of an understanding of the relationship between the transition from localized to delocalized excitation states and the nature of energy transfer in the liquid,

(e) Development of a more realistic theory in which the simplifications of the Drude model of the molecule and other approximations are removed.

It is my opinion that we are at the threshold of a vast expansion of our understanding of the electronic properties of liquids and other disordered systems. So little is known, either from experiment

or theory that almost any effort is likely to be rewarded with unexpected results. This field, unlike others, requires, more than anything else, new concepts and constructs and novel ways of interpreting strongly coupled phenomena. For some time it is likely that only qualitative interpretations of data will be possible, but with the steady accumulation of information and the creation of new interpretations we may look forward to the development of a quantitative theory encompassing in its scope the description of the properties of both ordered and disordered systems. Given that many systems of interest, including essentially all biological systems, are disordered to some extent, the importance of a deeper, broader, and more comprehensive description of the electronic properties of disordered systems cannot be over-estimated.

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